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Review of the National Ambient Air Quality Standards for Particulate Matter: Assessment of Scientific and Technical Information

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16. ABSTRACT

This paper evaluates and interprets the available scientific and technical information that the EPA staff believes is most relevant to the review of primary (health) and secondary (welfare) National Ambient Air Quality Standards for Particulate Matter (PM) and presents staff recommendations on alternative approaches to revising the standards. The assessment is intended to bridge the gap between the scientific review in the EPA criteria document for particulate matter and sulfur oxides and the judgements required of the Administrator in setting ambient air quality standards for particulate matter.

The major recommendations of the staff paper include the following:

1) that TSP be replaced by a new particle indicator that includes only those particles less than a nominal 10 μm (thoracic particles or PM_{10});

2) that the levels of both 24-hour and annual standards be revised;

3) that the statistical form of the standard should be changed;

4) consideration of a fine particle (<2.5 μm) secondary standard recognizing the advantages of combining any action with later decisions on acid deposition programs; and

5) consideration of secondary standards related to soiling and nuisance.

less than

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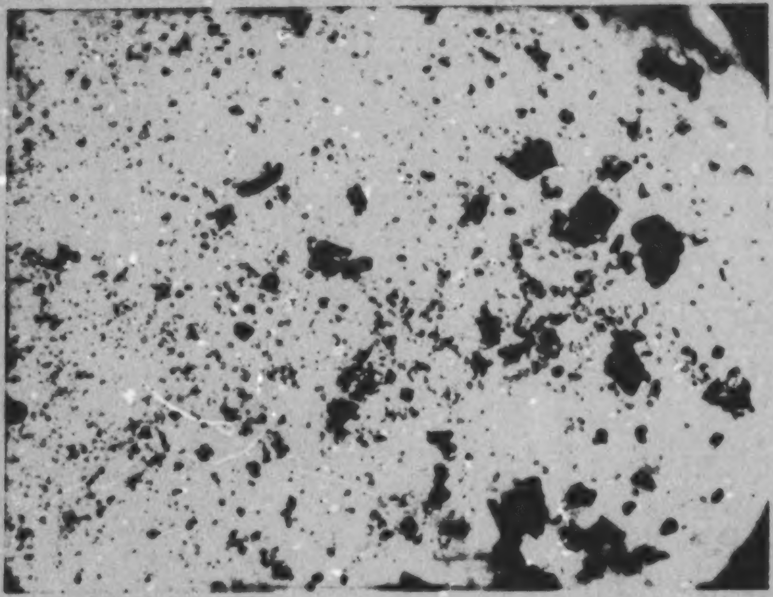
OAQPS Staff Paper



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Office of Air Quality Planning and Standards
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The cover illustration is a photomicrograph of an ambient particle sample collected in Chicago. Identifiable particles include calcite, oil soot, coal fragments, and soil particles. At this magnification (163X), submicrometer particles such as sulfate or carbon containing aerosols are not visible. Photo credit, Dr. Ron Draftz.

REVIEW OF THE NATIONAL AMBIENT AIR QUALITY STANDARDS FOR PARTICULATE MATTER: ASSESSMENT OF SCIENTIFIC AND TECHNICAL INFORMATION

OAQPS STAFF PAPER

Strategies and Air Standards Division
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January, 1982

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This staff paper is the product of the Office of Air Quality Planning and Standards (OAQPS). The principal authors include John Bachmann, Larry Zaragoza, Jeff Cohen, and John Haines. The report includes comments from OAQPS, the Office of Research and Development, and the Office of General Counsel within EPA and was formally reviewed by the Clean Air Scientific Advisory Committee.

Helpful comments and suggestions were also submitted by a number of independent scientists, by officials from several state agencies (California, New York, Arizona and Massachusetts), and by environmental and industrial groups including the National Resources Defense Council, the National Audubon Society, the Sierra Club, the American Iron and Steel Institute, the American Mining Congress, the American Petroleum Institute, the Utility Air Regulatory Group, and the Non-Ferrous Smelter Companies.

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EXECUTIVE SUMMARY

This paper evaluates and interprets the available scientific and technical information that the EPA staff believes is most relevant to the review of primary (health) and secondary (welfare) National Ambient Air Quality Standards (NAAQS) for particulate matter and presents staff recommendations on alternative approaches to revising the standards. Review of the NAAQS is a periodic process instituted to ensure the scientific adequacy of air quality standards and is required by Section 109 of the 1977 Clean Air Act Amendments. The assessment in this staff paper is intended to help bridge the gap between the scientific review contained in the EPA criteria document "Air Quality Criteria for Particulate Matter and Sulfur Oxides" and the judgments required of the Administrator in setting ambient standards for particulate matter. The staff paper is, therefore, an important element in the standards review process and provides an opportunity for public comment on proposed staff recommendations before they are presented to the Administrator.

Particulate matter represents a broad class of chemically and physically diverse substances that exist as discrete particles (liquid droplets or solids) ranging in size from molecular clusters of about 0.005 micrometers (μm) to coarse dusts on the order of 100 μm. Particles originate from a variety of stationary and mobile sources and may be emitted directly or formed in the atmosphere by transformations of gaseous emissions such as SO_2. The major chemical and physical properties of particulate matter vary greatly with time, region, meteorology and source category, complicating the assessment of health and welfare effects as related to various indicators of particulate pollution. Typical particle distributions reveal differences in origin and composition for fine particles (< 2.5 μm) and coarse particles (> 2.5 μm). The reference method for the current standards for particulate matter is the "hi volume" sampler which collects particulate matter of particle sizes up to 25-45 μm (so called "Total Suspended Particulate" or TSP).

At elevated concentrations, particulate matter can adversely affect human health, visibility, climate, materials, economic values, personal comfort and well-being, and vegetation. Components of particulate matter (e.g., sulfuric acid) also contribute to acid deposition. Typical long-term average levels of TSP range from 20-40 $\mu g/m^3$ in rural areas to over 150 $\mu g/m^3$ in the most polluted urban industrial areas. Maximum 24-hour TSP concentrations exceed 500 $\mu g/m^3$. Long-term fine particle (< 2.5 μm) levels range from 2 to 5 $\mu g/m^3$ in isolated arid western areas to 20-25 $\mu g/m^3$ in the rural East. The highest annual fine particle levels, on the order of 50 $\mu g/m^3$, occur in the most polluted urban industrial areas.

Primary Standards

The staff has reviewed scientific and technical information on the known and potential health effects of particulate matter cited in the criteria document. The information includes studies of respiratory tract deposition of particles, studies of mechanisms of toxicity, effects of high exposures to various particulate substances in controlled human and animal studies, epidemiological studies, and air quality information. Based on this review, the staff derives the following conclusions.

1) The mechanisms by which inhaled particles may pose health risks involve (a) penetration into and deposition of particles in the various regions of the respiratory tract, and (b) the biological responses to the deposited materials.

2) The risks of adverse effects associated with deposition of ambient fine and coarse particles in the thorax (tracheobronchial and

alveolar regions of the respiratory tract) are markedly greater than for deposition in the extrathoracic (head) region. Maximum particle penetration to the thoracic regions occurs during oronasal or mouth breathing.

3) The major effects categories of concern associated with high exposures to particulate matter include: (a) effects on respiratory mechanics and symptoms, (b) aggravation of existing respiratory and cardiovascular disease, (c) effects on clearance and other host defense mechanisms, (d) morphological alterations, (e) carcinogenesis, and (f) mortality.

4) The major subgroups of the population that appear likely to be most sensitive to the effects of particulate matter include: (a) individuals with chronic obstructive pulmonary or cardiovascular disease, (b) individuals with influenza, (c) asthmatics, (d) the elderly, (e) children, (f) smokers, and (g) mouth or oronasal breathers.

5) Although controlled animal and human studies, and qualitative epidemiological results can provide important insights into the health risks from particles, the most useful concentration-response information comes from a limited set of community epidemiological studies conducted over the last 25 years in Great Britain and the United States.

Based on the scientific and technical reviews as well as policy considerations, the staff makes the following recommendations with respect to primary particulate matter standards.

1) Despite the variability in the composition of ambient particles with time and space, the available data suggest that reductions in ambient particulate matter in Great Britain and the U.S. have

benefited public health and reduced the need for separate control programs for many of the more innately toxic components of particulate matter. Elevated particulate matter exposures in current U.S. settings most frequently occur without concomitant high SO_2 levels. Considering these observations, a separate general particulate matter standard remains a reasonable public health policy choice.

2) The current TSP standard directs control efforts towards particles of lower risk to health because of its inclusion of larger particles which can dominate the measured mass concentration, but which are deposited only in the extrathoracic region. A new particle indicator representing those particles capable of penetrating the thoracic regions (thoracic particles, TP) is recommended. Protection of sensitive individuals breathing oronasally or by mouth, sampler reliability, and the convention recently adopted by the International Standards Organization (ISO) suggest that the particle size range include those particles less than a nominal 10 μm (D_{50}). Sampler performance criteria should be related to respiratory tract deposition data. Prototype samplers meeting these criteria are being field tested; reliable commercially available models must await test results.

3) Both short-term (24-hour) and annual arithmetic mean standards are recommended. The short-term standard should be expressed in statistical form with the decision on the allowable number of exceedances made in conjunction with establishing a level for the standard.

4) Selecting a level for a particulate matter standard with an adequate margin of safety will involve a number of uncertainties

> in addition to those involved in making judgments on health risks associated with specific substances such as CO or SO_2. Quantitative assessments must be based on limited epidemiological studies conducted in times and places where pollutant composition may have varied considerably from current U.S. atmospheres. Epidemiological studies are generally subject to a number of inherent difficulties involving confounding variables and somewhat limited sensitivity. Most studies have used British smoke (a pseudo mass indicator related to small particle (< 4.5 μm) darkness) or TSP as particle indicators. None of the published studies have used the recommended TP (< 10 μm) indicator. Thus, appropriate assumptions must be used to express available results in common units.

The staff assessment of short-term epidemiological data is summarized in Table 1; levels are expressed in both the original and TP units. The "effects likely" row denotes concentration ranges derived from the criteria document at or above which there appears greatest certainty that effects would occur. The data do not, however, show evidence of clear population thresholds but suggest a continuum of response with both the risk of effects occurring and the magnitude of any potential effect decreasing with concentration. Thus, effects may be possible at levels below those listed in the "effects likely" row, but the evidence and risks at lower levels are much less certain.

Based on this staff assessment, the range of 24-hour TP levels of interest are 150 to 350 $\mu g/m^3$. Under the conditions prevailing during the London studies, the upper end of the range represents levels at which effects are likely in the sensitive populations studied. Given the uncertainties in translating these results to U.S. conditions and the seriousness

TABLE 1. STAFF ASSESSMENT OF SHORT-TERM EPIDEMIOLOGICAL STUDIES

	Measured British Smoke Levels (as $\mu g/m^3$)			Equivalent TP ($<10\ \mu m$) Levels ($\mu g/m^3$)
Effects/ Study	Daily Mortality in London[1]	Aggravation of Bronchitis[2]	Combined Range	Combined Range[3]
Effects Likely	500-1000	250*-500*	250-500	350-600
Effects Possible	150*-500	<250*	150-250	150-350

*Indicates levels used for upper or lower bound of range.

[1] Deviations in daily mortality from mean levels examined in 3 studies encompassing individual London winters of 1958-59 and 14 aggregate winters from 1958-72. Early winters were dominated by high smoke dominated by coal combustion emissions and high SO_2 with frequent fogs.

[2] Examination of symptoms reported by bronchitics in London. Studies conducted from the mid-1950's to the early 1970's.

[3] Conversion assumes that for London smoke conditions, $BS \leq TP \leq TSP$. Precise conversions are not possible. The lower bound assumption ($BS = TP$) incorporates some margin of safety and is used to estimate the lower bound in the effects possible range. The upper bound ($TP = TSP = BS + 100\ \mu g/m^3$) likely overestimates TP levels and is used to provide estimates of levels where effects are most likely.

of the potential health effects, the upper end of the above range contains no identifiable margin of safety and should not be considered as an appropriate standard alternative. The uncertainties and the nature of the potential effects are important margin-of-safety considerations. Neither the studies summarized above nor more qualitative studies of effects in other sensitive population groups (e.g., asthmatics, children), or effects in controlled human or animal stuides provide scientific support for health risks of consequence below 150 $\mu g/m^3$. These qualitative data as well as factors such as aerosol composition and exposure characteristics should also be considered in evaluating margins of safety associated with alternative standards in the range of 150 $\mu g/m^3$ to something below 350 $\mu g/m^3$.

The staff assessment of important long-term epidemiological data is summarized in Table 2. Long-term epidemiological studies are subject to additional confounding variables that reduce their sensitivity and make interpretation more difficult. The "effects likely" levels are derived from the criteria document, but again, no clear population thresholds exist for all effects indicators. Some risk of effects are possible at lower levels, but these are uncertain and difficult to detect in these studies.

Based on this staff assessment, the range of annual TP levels of interest are 55 to 110 $\mu g/m^3$. The upper end of this range overlaps the somewhat uncertain "effects levels" derived from these studies. Due to these uncertainties, the upper end of the range (110 $\mu g/m^3$) may not include any margin of safety, and should not be considered as an appropriate standard alternative. The lower end (55 $\mu g/m^3$) represents a level where some risk of symptomatic effects might remain but no detectable differences in pulmonary function or marked increases in respiratory diseases are expected. Increases in symptomatic effects at the lower levels are uncertain and small in comparison to baseline rates.

TABLE 2. STAFF ASSESSMENT OF LONG-TERM EPIDEMIOLOGICAL STUDIES

	Measured BS Levels (as $\mu g/m^3$)	Measured TSP Levels			Equivalent TP Levels ($\mu g/m^3$)
Effects/Study	Increased Respiratory Disease, Reduced Lung Function in Children[1]	Increased Respiratory Disease Symptoms, Small Reduction in Lung Function in Adults[2]	Increased Respiratory Symptoms in Adults[3]	Combined Range	Combined Range[4]
Effects Likely	230-300 BS	180*	--	≥ 180	90-110
Effects Possible	< 230 BS	130-180*	60-150 (110*)	110-180	55-110
No Significant Effects Noted	--	80*-130	--	80-110	40-55

*Indicates levels used for upper or lower bound of range.

[1] Study conducted in 1963-65 in Sheffield, England. BS levels (as $\mu g/m^3$) uncertain.

[2] Studies conducted in 1961-73 in Berlin, NH. Major source in community was pulp mill. Effects level (180 $\mu g/m^3$) based on 2 month average. Effects on lung function were relatively small.

[3] Study conducted in 1973. Exposures reflect 1965-73 data in Ansonia, CT. Median value used as an indicator. Essentially negative study. No effects on lung function, but some suggestion of effects on respiratory symptoms.

[4] Conversion based on estimated ratio of TP/TSP for current (1980-81) U.S. atmospheres. The ratio ranges between about 0.5 to 0.6. These numbers are used as lower and upper bounds for estimating TP equivalents from TSP values.

When evaluating margins of safety for an annual standard, it is particularly important to examine the results of qualitative data from a number of epidemiological, animal, and air quality studies. These suggest concern for effects not directly evaluted in the studies listed in Table 2. Such effects include damage to lung tissues contributing to chronic respiratory disease, cancer, and premature mortality. The available scientific data do not suggest major risks for these effects categories at current ambient particle levels in most U.S. areas. Nevertheless, the risk that both fine and coarse particles may produce these responses supports the need to limit long-term levels of TP for a variety of aerosol compositions.

Because of different form, averaging procedure and size range, precise comparisions between the above ranges of TP standards and the current primary TSP standards are not possible.* The lower bounds, taken together, result in standards roughly equivalent in stringency to the current standards. In general, the rest of the ranges represent increasing degrees of relaxation as compared with the current standards. At the lower concentrations in the ranges, much of the relaxation would result because only smaller particle sizes would be collected. Thus, a city where exceedance of the TSP standard was largely dominated by coarse mode dust (with substantial mass of particles greater than 10 μm) would be less likely to violate a comparable TP standard than would an area where exceedance of the TSP standard was dominated by particles smaller than 10 μm. At higher concentrations in the above ranges, standards would permit increased levels for TP as well as for larger particles.

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*By applying observed TP/TSP ratios and other factors, crude comparisons can be made. The current annual TSP standard (75 μg/m^3, geometric mean) is roughly equivalent to an arithmetic mean of 50 μg/m^3 as TP. The numerical value of the 24 hr TSP standard (260 μg/m^3) is roughly equivalent to 140 μg/m^3 TP, but this does not account for differences between the deterministic (current standard) and recommended statistical form.

Secondary Standards

The staff examined information in the criteria document relevant to the review of the secondary standards. Categories of welfare effects examined include visibility and climate, man-made materials, vegetation, and personal comfort and well-being. Major staff conclusions and recommendations are summarized below.

1) a) Impairment of visibility by fine particles over urban to multistate regions clearly affects public welfare. Fine particles or major constituents thereof also are implicated in climatic effects, materials damage, soiling, and acid deposition. Neither the current secondary TSP standard nor the recommended ranges of TP standards will protect visibility in an effective manner. The staff, therefore, recommends consideration of a fine particle secondary standard, based primarily on the relatively well-defined quantitative relationships between fine mass and visibility.

b) If a fine particle standard is selected, a seasonal (calendar quarter) averaging time could provide a statistically stable target and yet achieve most short or long-term visibility goals. Consideration should be given to specifying a spatial average of three or more monitors placed at distances on the order of 16-50 km.

c) Despite the fact that the public is concerned about visibility and is willing to pay something for clean air, quantitative bases for evaluating visibility goals have not been established. Therefore, the level of any standard must be based on the judgment of the Administrator after consideration of aesthetics and transportation, as well as non-visibility related effects. The staff recommends that any national standards focus on welfare effects associated with multistate eastern regional (and western urban) haze. Such standards would not of themselves protect sensitive scenic areas of the West, but these areas are directly and indirectly addressed by other provisions of the Clean Air Act.

d) Empirical ranges for standards can be derived from approximate estimates of eastern natural background and current summertime fine particle levels. The range thus derived is 8-25 $\mu g/m^3$, seasonal and spatial average. The upper portion of the range would tend to maintain the *status quo* in the East. Current summertime visual range in much of the East is about 9-15 miles. Because the lower portion of the range approaches natural background levels, standards set at the lower levels would be, in all practicality, unattainable in most of the eastern U.S. Estimated summertime visibility under eastern natural background conditions is on the order of 3 to 5 times greater than under current conditions.

e) Because regional fine particles in the East appear to be influenced most strongly by sulfates, adoption of a fine particle standard would trigger a substantial departure from current approaches to particle control strategies. The evidence suggests that multistate control of regional sulfur oxide emissons might be needed to reduce fine particle levels. Thus, fine particle/visibility-climate effects are linked to acid deposition, and these problems would likely be ameliorated by similar control strategies. Addressing these welfare effects with a common standard or control strategy is likely to be more efficient than establishing separate control approaches for each. Appropriate scientifically based targets and control strategies for acid deposition are not yet available.

2) Although potential effects on climate support the consideration of a fine particle standard, quantitative relationships are not well enough developed to provide the principal basis for selecting the level of the standard.

3) Consideration should be given to soiling and nuisance effects in determining whether a secondary standard for TP or for TSP or some other large particle indicator is desirable to supplement the primary health and secondary fine particle standards. The available data base on such effects is, however, largely qualitative. Therefore, the basis for selecting a particular level for a secondary TP or TSP standard is a matter of judgment.

4) While chemically active fine mode and hygroscopic coarse mode particles have been qualitatively associated with materials damage, the available data do not clearly suggest major effects of particles on materials for concentrations at or below the ranges recommended for the primary health and secondary visibility standards. Therefore, a secondary standard based solely on materials damage is not recommended.

5) The staff concludes that a secondary particle standard is not needed to protect vegetation.

6) The acid deposition issue will not be addressed directly in the review of the particulate matter standards.

REVIEW OF THE NATIONAL AMBIENT AIR QUALITY STANDARDS FOR PARTICULATE MATTER: ASSESSMENT OF SCIENTIFIC AND TECHNICAL INFORMATION

OAQPS STAFF PAPER

I. PURPOSE

This paper evaluates and interprets the most relevant scientific and technical information reviewed in the draft EPA document "Air Quality Criteria for Particulate Matter and Sulfur Oxides" (EPA, 1981) in order to better specify the critical elements which EPA staff believes should be considered in the possible revision of the primary and secondary particulate matter National Ambient Air Quality Standards (NAAQS). This assessment is intended to help bridge the gap between the scientific review contained in the criteria document and the judgments required of the Administrator in setting ambient standards for particulate matter. As such, particular emphasis is placed on identifying those conclusions and uncertainties in the available scientific literature that the staff believes should be considered in selecting a particulate pollutant indicator, form, and level for the primary standards. While the paper should be of use to all parties interested in the standards review, it is written for those decision makers, scientists, and staff who have some familiarity with the technical discussions contained in the criteria document.

II. BACKGROUND

Since 1970 the Clean Air Act as amended has provided authority and guidance for the listing of certain ambient air pollutants which may endanger public health or welfare and the setting and revising of NAAQS for those pollutants. Primary standards must be based on health effects criteria and provide an adequate margin of safety to ensure protection of public health. As several recent judicial decisions have made clear, the economic and technological feasibility of attaining primary standards are not to be considered in setting them, although such factors may be considered to a degree in the development of state plans to implement the standards (D.C. Cir., 1980, 1981). Further guidance provided in the legislative history of the Act indicates that the standards should be set at "the maximum permissible ambient air level . . . which will protect the health of any (sensitive) group of the population." Also, margins of safety are to be provided such that the standards will afford "a reasonable degree of protection . . . against hazards which research has not yet identified." (Committee on Public Works, 1974). In the final analysis, the EPA Administrator must make a policy decision in setting the primary standard, based on her judgment regarding the implications of all the health effects evidence and the requirement that an adequate margin of safety be provided.

Secondary ambient air quality standards must be adequate to protect the public welfare from any known or anticipated adverse effects associated with the presence of a listed ambient air pollutant. Welfare effects, which are defined in section 302(h) of the Act, include effects on vegetation, visibility, water, crops, man-made materials, animals, economic values and personal comfort and well-being. In specifying a level or levels for secondary standards the Administrator must determine at which point the effects become "adverse" and base her judgment on the welfare effects criteria.

The current primary standards for particulate matter (to protect public health) are 75 micrograms per cubic meter ($\mu g/m^3$) annual geometric mean, and 260 $\mu g/m^3$, maximum 24 hour concentration not to be exceeded more than once per year. The current secondary standard for particulate matter (to protect public welfare) is 150 $\mu g/m^3$, maximum 24 hour concentration, not to be exceeded more than once per year. In addition, the secondary standard specifies a 60 $\mu g/m^3$ annual geometric mean guide for the achievement of the 24-hour standard. The reference method for measuring attainment of both the primary and secondary standards is the "hi-volume" sampler (40 CFR Part 50, Appendix B), which effectively collects particulate matter in the range of up to 25-45 micrometers (μm) (so-called "total suspended particulate," or "TSP"). Thus, TSP is the current indicator for the particulate matter standards.

Preliminary drafts of this paper were reviewed by the Clean Air Scientific Advisory Committee (CASAC) in July and November, 1981. This final product incorporates the suggestions and recommendations of the CASAC as well as other appropriate comments received on the initial drafts. The CASAC closure memo on the staff paper (Friedlander, 1982) is reprinted in Appendix E.

III. APPROACH

The approach used in this paper is to assess and integrate information derived from the criteria review in the context of those critical elements which the staff believes should be considered in the review of the primary and secondary standards. Particular attention is drawn to those judgments that must be based on the careful interpretation of incomplete or uncertain evidence. In such instances, the paper states the staff's evaluation of the evidence as it relates to a specific judgment, sets forth appropriate alternatives that should be considered, and recommends a course of action.

Sections IV and V review and integrate important scientific and technical information relevant to standard setting. Because of the complex nature of particulate pollution, this review is unusually lengthy. Therefore, much of this material is included in appendices (A and B) with only summaries in the main body of the paper. Section IV presents relevant features of historical and current U.S. air quality to support discussions of both primary and secondary standards. Section V addresses the essential elements with regard to the primary standards; these include the following:

1) identification of possible mechanisms of toxicity;
2) description of effects and judgment of critical effects of concern for standard setting;
3) identification of most sensitive population groups; and
4) discussion of community studies relating level(s) and duration(s) of exposure to indicators of health effects.

Drawing from the discussion in Sections IV and V, Section VI identifies and assesses the factors the staff believes should be considered in selecting a particulate pollutant indicator, form, and level of primary standards. Preliminary staff recommendations on alternative policy options in each of these areas also are presented.

Section VII examines information in the criteria document the staff believes is most relevant with respect to secondary standards and focuses on the effects of particulate matter on visibility and climate, man-made materials, vegetation, and personal comfort and well-being. Some of the scientific and technical review is included in Appendix C. The elements addressed include:

1) description of effects and judgment of the critical effects of concern for standard setting;
2) identification of causal mechanisms;
3) studies relating level(s) and duration(s) of exposure to indicators of effects; and

4) factors to be considered in selecting a particulate pollutant indicator, form, and level of secondary standards.

Preliminary staff recommendations on policy options for secondary standards are also presented.

IV. AIR QUALITY CONSIDERATIONS

More than any other criteria pollutant, "particulate matter" represents a broad class of chemically and physically diverse substances. Their principal common feature is existence as discrete particles in the condensed (liquid or solid) phase ranging in size from molecular clusters of 0.005 μm to coarse particles on the order of 100 μm.* The major chemical and physical properties of particulate matter vary greatly with time, region, meteorology, and source category. It is to be expected, then, that the effects of given quantities of particles on public health and welfare also will vary. This variable composition complicates the evaluation of the applicability of specific particle health and welfare studies for establishing National Ambient Air Quality Standards. This section of the staff paper summarizes some key features of our understanding of historical and current particulate matter composition to provide perspective for subsequent interpretation of health and welfare effects studies.

The historical aerosol composition information is intended to assist in interpreting the relevance of historical health effects studies to current U.S. atmospheres. Many of the more important epidemiological studies were conducted in Great Britain, particularly in London, and in New York City in the 1950's and 60's. Important aspects of current physical and chemical distribution of aerosols are presented to support identification of those aerosol fractions potentially responsible for health and welfare effects and evaluation of standard levels.

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*Where not otherwise specified, particle sizes reported in this paper reflect aerodynamic equivalent diameter (AED). A number of terms (e.g., fine, coarse, inhalable, thoracic, TSP) are used to describe various functions of particulate matter. Many of these terms are defined by the instruments used for measurement. The major particle indicators discussed in this paper are defined in Appendix D.

A. Historical Trends

1. London 1950's - 1960's

Although implementation of the British Clean Air Act effected dramatic air quality improvements from 1956 to 1970, particle and sulfur oxide levels from 1955 to 1963 were remarkably high by present day standards. Typical annual SO_2 and smoke (a pseudo-mass indicator related to small particle (< 5 μm) darkness, McFarland et al., 1981) concentrations ranged between 200 to over 300 $\mu g/m^3$ in Metropolitan London from 1955-1960 (WSL, 1967). Details on short and long-term levels of interest are reported in the later health effects discussions. The major source of these pollutants was the inefficient combustion of fuel, particularly domestic coal. In addition, "industrial processes, building and demolition, wear of roads, tyres (sic) and other materials, the burning of vehicle fuels containing additives" contributed to particulate loadings (Lawther et al., 1968). One would expect atmospheres dominated by incomplete coal combustion emissions to contain large amounts of carbonaceous material ranging from fine soot to coarse cinders, various inorganic elements (e.g., mercury, arsenic, zinc) found in coal, primary sulfuric acid aerosol as well as SO_2, and particularly in the case of British coal, chlorine containing compounds such as gaseous hydrochloric acid.

Unfortunately, actual ambient measurements of expected components are rare. The best bench-mark for evaluating the composition of London particles (and comparing them to contemporary U.S. aerosols) is found in a series of investigations conducted by Waller, Lawther, Commins and other air pollution researchers with the Medical Research Council at the St. Bartholomew's Hospital Medical College in London. This location in the "City," a one square mile area encompassing the business district of London, may not be representative of greater London levels, but should reflect general composition and trends. The monitors were fairly elevated, generally 22 m or more above ground level. SO_2 levels appear 50 to 100 $\mu g/m^3$ higher than levels reported elsewhere in London, but smoke readings were 50 to 100 $\mu g/m^3$ lower (Commins and Waller, 1967; WSL, 1967). The authors suggest that their site may be strongly influenced by local point sources of SO_2, making SO_2 levels and trends atypical (Commins and Waller, 1967). The reason for lower smoke readings is less clear and could involve lower amounts of nearby domestic coal use, since the site was in a smoke control zone; the lower readings also might be due in part to the more reliable calibration of the smoke readings with actual mass concentrations as performed by the St. Bartholomew's group (Waller, 1964).

The investigators operated standard U.S. hi-volume air samplers (particle size range collected usually less than about 25-45 μm) as well as conducting special size and composition studies over the eight year period from 1955-1963. TSP readings are plotted in Figure 4-1. Contemporary readings from some New York City sites (Eisenbud, 1980) are shown for comparison. The beneficial effects of pollution control programs are apparent from the trends. Although analyses of the composition of London TSP samples were performed, long-term composition data are regrettably available only for the benzene soluble organics (BSO) fraction (see Figure 4-1). This fraction contains polycyclic organics (POM) as well as other non-polar organic aerosols. On average, about 16% of collected TSP mass consisted of this organic material, but some individual samples taken during periods of high pollution contained more than 50% BSO (Commins and Waller, 1967). Benzo(a)pyrene (BaP), often used as an indicator of

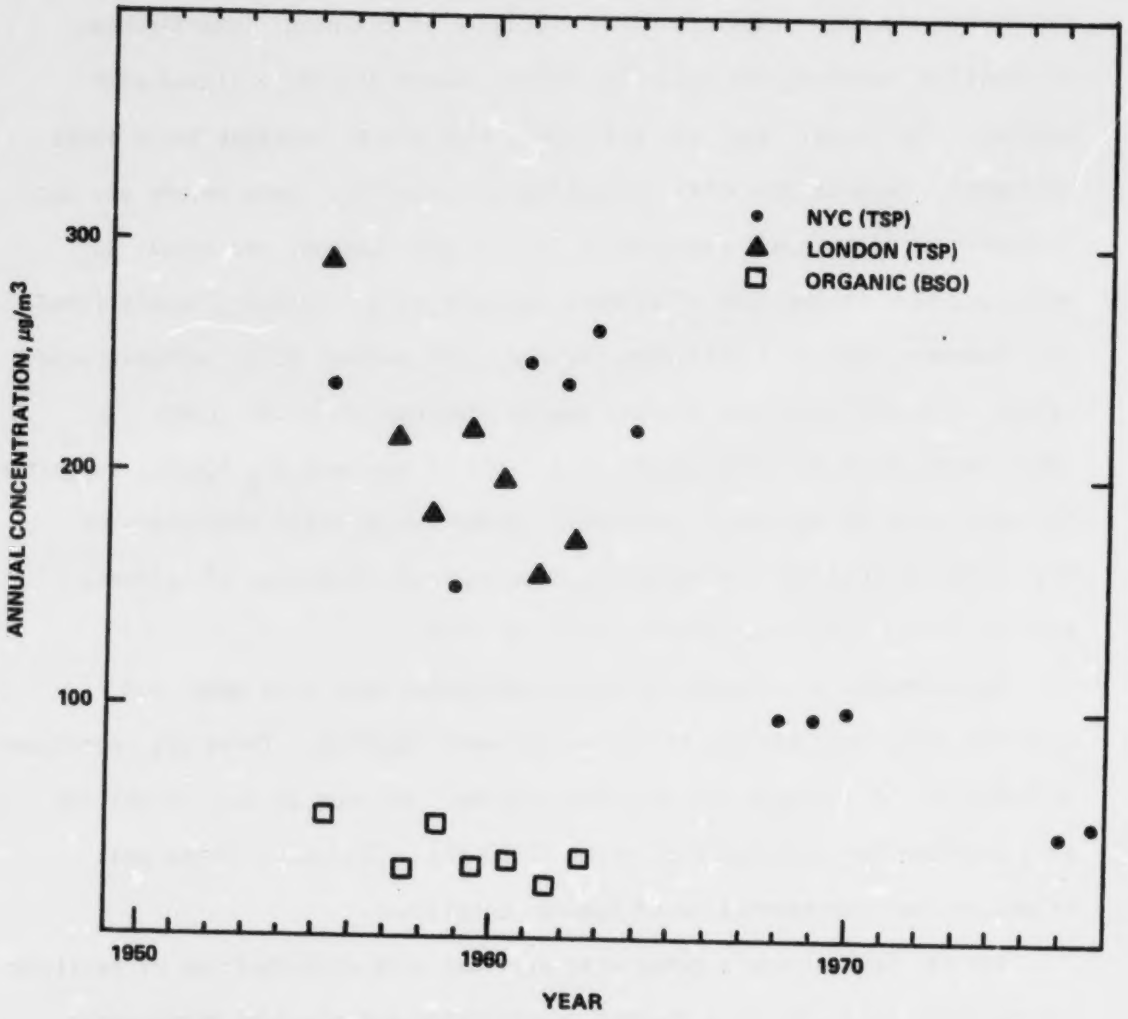

Figure 4-1. Trends in TSP and benzene soluble organics at a central London site (Commins and Waller, 1967), and TSP trends for New York City as summarized by Eisenbud (1980). TSP in both cities were considerably higher than present day levels in most urban areas.

carcinogenic POM, typically ranged from 2-30 ng/m^3 with maximal peaks on the order of 1 μg/m^3.

Some annual (1964-65) and episodic data for London also are available for strong acids (reported as sulfuric acid) as measured by filtration and subsequent titration (Waller, 1963). Because of potential interferences in sampling, handling and analysis, these numbers should be viewed with caution. The annual level was 5.4 μg/m^3, with winter averages twice those of summer. Despite potential methodological problems, these values are not inconsistent with levels expected if 1-2% of the seasonal and annual SO_x emissions were in the form of primary sulfuric acid. Episodic levels (1957-64), however, present a different picture, with maximal daily sulfuric acid values of 30-350 μg/m^3 and maximal hourly readings of 30-680 μg/m^3. In these cases, sulfuric acid values were 5-10% of measured SO_2 levels, suggesting the occurrence of secondary formation. Under the episodic conditions of high particle loadings and humidity, heterogeneous formation of sulfuric acid is likely (Criteria Document (CD), p. 2-33).

Measurements of a number of other substances were also made, but unfortunately, only maximum hourly values were reported. These are reproduced in Table 4-1 to indicate the relative ranking. Maximum 24 hour values are also provided for selected U.S. sites (1960-65). Obviously, these peak values are not representative of typical conditions.

The St. Bartholomew's group also examined size distributions of particles and sulfuric acid. Using a thermal precipitator and electron microscopy, Waller _et al_. (1963) found numerous single particles smaller than 1 μm, aggregates up to 1 - 2 μm, and evidence of partially neutralized sulfuric acid droplets.

TABLE 4-1. COMPARISON OF MEASURED COMPONENTS OF TSP IN U.S. CITIES (1960-65) WITH MAXIMUM 1-HOUR VALUES IN LONDON (1955-63)

	UNITED STATES [a]			LONDON [b]
		Concentration μg/m^3		
Pollutant	Number of Stations	Arith. average	Maximum 24-Hour	Maximum 1-Hour (1 site)
Suspended Particles	291	105	1254 (TSP)	9700 (Smoke)[c]
Fractions:				
Benzene-soluble organics	218	6.8	—	—
Chloride (water soluble)	—	—	—	410
Nitrates	96	2.6	39.7	5
Sulfates	96	10.6	101.2	666
Sulfuric acid	—	—	—	680
Ammonium	56	1.3	75.5	—
Antimony	35	0.001	0.160	<1
Arsenic	133	0.02	—	—
Beryllium	100	<0.0005	0.010	<1
Bismuth	35	<0.0005	0.064	<1
Cadmium	35	0.002	0.420	1
Calcium	—	—	—	82
Chromium	103	0.015	0.330	2
Cobalt	35	<0.0005	0.060	<1
Copper	103	0.09	10.00	2
Iron	104	1.58	22.00	25
Lead	104	0.79	8.60	22
Manganese	103	0.10	9.98	5
Molybdenum	35	<0.005	0.78	<1
Nickel	103	0.034	0.460	1
Tin	85	0.02	0.50	2
Titanium	104	0.04	1.10	1
Vanadium	99	0.050	2.200	2
Zinc	99	0.67	58.00	24
Gross beta radioactivity	323	(0.8 pCi/m^3)	(12.4 pCi/m^3)	—

[a] U.S. HEW, 1969.

[b] Commins and Waller, 1967.

[c] British Smoke (BS) data reported as μg/m^3. These data are not directly comparable with TSP.

The aggregate, which varied with source area, apparently consisted of carbonaceous material (carbon and organics) from incomplete combustion. The mass median diameter (MMD) was 0.5 - 1 μm in normal weather and somewhat higher (but indeterminate) in foggy weather.

These values may underestimate the true mass median diameter* of the total aerosol. Due to the nature of the thermal precipitator (Hodkinson, 1960), larger particles (> 2-5 μm) may settle out or be inefficiently moved to the collection plate. The fact that the estimated mass from the microscopy determination generally approximated smoke readings (Waller et al., 1963), tends to support the notion that only the smaller portion of the aerosol was actually examined. At this site, calibrated smoke readings were consistently about 100 $\mu g/m^3$ below TSP levels, particularly at lower concentrations ($< 500\ \mu g/m^3$) (Commins and Waller, 1967). Moreover, samples collected at the elevation of the St. Bartholomew's site (22 m) would not be representative of ground level large particle concentrations. Other considerations, notably measurements of dry deposition and the dispersed nature of coal and oil emissions (which include large particles), support the notion that substantial quantities of large particles (2 to over 30 μm) were emitted and were probably present in the ambient air of British cities (Carey, 1959). Nevertheless, during extreme episodes ($BS > 500\ \mu g/m^3$), with low windspeeds, large particles would be expected to settle relatively rapidly near source areas.

Waller (1963) estimated the size distribution of sulfuric acid droplets ($pH < 2$) using a cascade impactor coated with a pH indicator. At lower humidities, the mass median diameter of these droplets was less than 0.5 μm. During an episode with higher humidities (∿ 85%), the mass median diameter was 0.5 μm, but 16% of the sulfuric acid mass was greater than 4.0 μm (estimated σ_g of 2.1). During high humidity fogs measurable amounts of large (20 μm) sulfuric acid droplets (pH < 2.0) were often observed.

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*MMD is used as an indicator of relative particle size of a lognormally distributed aerosol. The term is used loosely here, since the distribution may not have been lognormal.

2. U.S. Particle Trends

As indicated in Figure 4-1, annual TSP levels from 1955-65 in New York City were in the same range as those in London. However, even during this period, the influence of particles from inefficient fuel combustion may have been somewhat lower than for London. Organics (BSO) from 1961-65 were about 10% or less of total particulate mass as compared to 15-20% at the London site (DHEW, 1969). Long-term and maximum daily values of a number of compounds for a number of U.S. cities are compared with maximum hourly London data in Table 4-1. Although hourly values cannot be strictly compared to 24 hour data, maximum levels for sulfates, lead, and some trace elements were probably higher in London. Maximal levels of nitrates, manganese, and some additional elements were higher in U.S. cities. A striking feature of available information in both London and the U.S. is the inability to account for more than about 25% of TSP by the available compositional analyses.

From the early 1960's through the mid-1970's, U.S. control programs continued to result in substantial reductions in TSP and SO_2 levels in most of the more polluted urban areas. In these areas, concentrations of a number of components of interest also decreased markedly, most notably carbonaceous material (BSO), trace elements (lead, nickel, vanadium, cadmium, and iron), and in some areas, sulfates (Faoro and McMullen, 1977; Faoro, 1975; Altshuller, 1976). During this same time period, available information suggests that eastern U.S. regional levels of summertime fine particles, particularly sulfates, increased (EPA, 1975b, 1979; Frank and Posseill, 1976; Altshuller, 1976).

B. Current U.S. Aerosols

Changes in the nature and distribution of combustion related and other sources, and the implementation of various control programs since the 1970's have brought about substantial changes in the distribution, quantity and composition of airborne particles in the U.S. Moreover, the quantity and nature of accompanying pollutant gases are also substantially different than those of the earlier London and U.S. atmospheres discussed above. Notably, areas or times with higher particle loadings are now more likely to be accompanied by lower SO_2 levels and higher oxidant levels than occurred in the past. Although the diversity of particles and gases in U.S. atmospheres is, if anything, greater, important advances in measurement and approach in aerosol science during the 1970's have afforded a far greater understanding of the general features and origins of contemporary airborne particles. The criteria document discusses the results of a number of these advances in chapters 2-6. Some key features, and their implications, are outlined below.

1. The Multimodal Distribution

The atmospheric aerosol surface area, volume, and mass tend to be distributed in two to three distinct size modes (Figure 4-2):

1) The nuclei (Aitken) mode (< 0.1 μm) accounts for most of the particles by number, some of the surface area, and little mass, but is short-lived and observed only near combustion sources. Through coagulation, nuclei mode particles grow into the next mode, the accumulation mode.

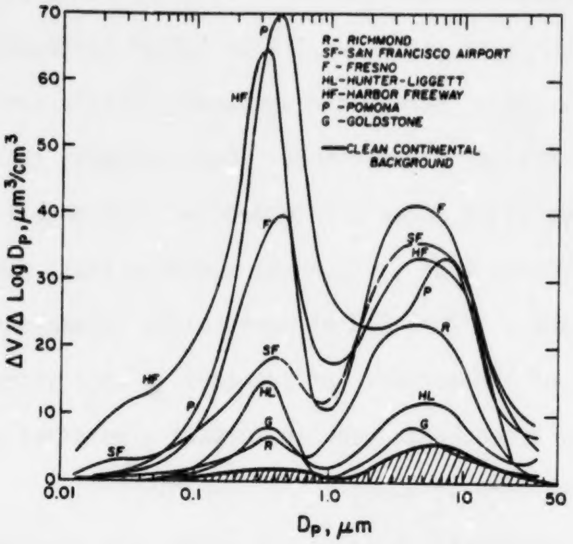


Figure 4-2. Grand average volume size distribution for seven sites in the California ACHEX study in 1972. Particle size reflects optical rather than aerodynamic diameter. Volume (and mass) tends to be bimodally distributed although the relative proportion of fine (<1-3 µm) and coarse (>1-3 µm) modes as well as location of maxima and minima vary considerably. The increased volume of 0.01 - 0.1 µm particles at the Harbor Freeway site is evidence of the transient nuclei mode near a strong source (automobiles) (Whitby, 1980).

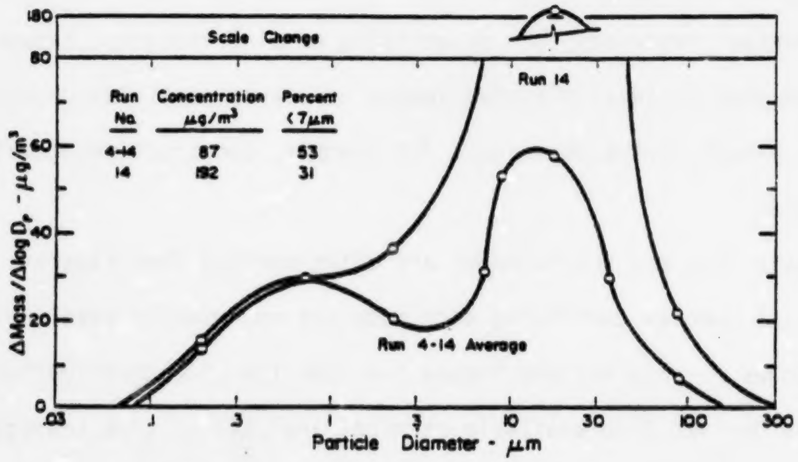


Figure 4-3. Bimodal mass-size distributions measured with impactors. Unlike Figure 4-2, particle size reflects aerodynamic diameter. The increase in coarse mode particles in Run 14 shows the influence of upwind construction activity. Note that the "minimum" in the original distribution disappears and the coarse increase occurs in the range of 1 to 300 µm. (Reprinted from NAS, 1977).

2) The accumulation mode (0.1 - 2.5 μm) contains much of the surface area and about 1/3 to 1/2 of particulate volume and mass. Under normal circumstances, little mass is transferred into the next larger mode. Collectively, the nuclei and accumulation modes are termed the fine mode (< 2.5 μm).

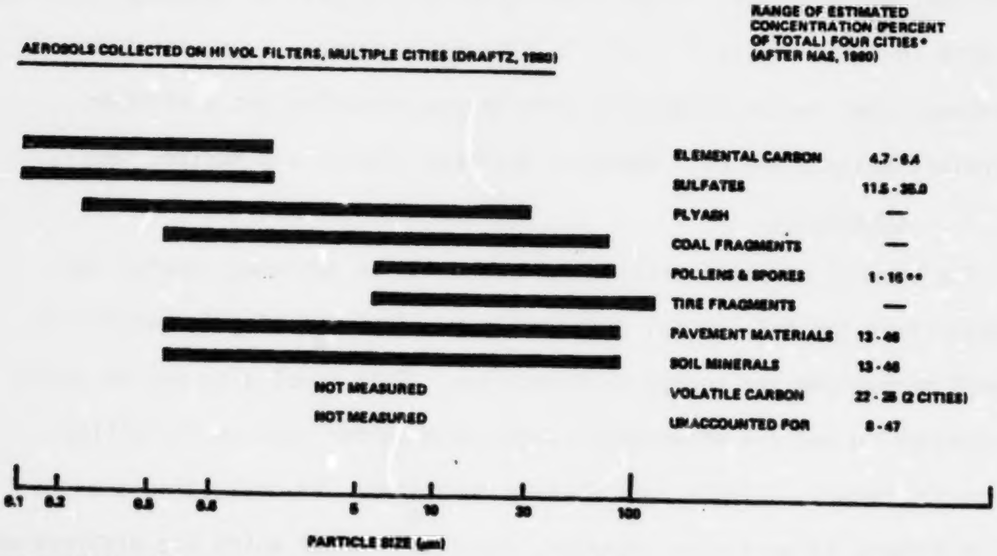
3) The coarse mode (> 2.5 μm) contains small numbers of particles and about 1/2 to 2/3 of particulate volume and mass. Although much of the aerosol surface area is associated with the fine mode, a secondary peak in surface area often appears in the coarse mode at about 10 μm (NAS, 1977a).

Although substantial overlap can exist, the fine and coarse modes tend to have more or less distinct origins, elemental distribution, residence times, and removal processes. Fine particles originate in the nuclei mode by condensation of materials produced during combustion (e.g., inorganic lead) or atmospheric transformation (e.g., sulfuric acid). Due to long residence times and atmospheric formation, fine particle levels can build up far from source regions over large geographical areas. Coarse particles are largely derived from mechanical processes such as grinding or wind erosion. Because they settle out more rapidly, elevated levels of coarse particles usually occur only near strong source emissions, for example, construction activity (Figure 4-3).

Because fine and coarse modes are often derived from separate mechanisms, the chemical species comprising each mode are not usually evenly distributed between modes. Table 4-2 and Figure 4-4 show the size distribution of various substances derived from available chemical analysis of size segregated samples and microscopy of hi-vol filters. Although regular elemental/chemical segregation occurs, each mode is chemically heterogeneous in the sense that each consists of a number of different substances. The dominant

TABLE 4-2. NORMAL SIZE RANGES FOR COMMONLY FOUND CHEMICALS, BASED ON IMPACTOR STUDIES
(After Miller *et al.*, 1979)

Normally fine	Normally coarse	Normally bimodal	Variable
Sulfates	Iron, calcium	Chloride	Nickel
Organic (condensed vapors)	Titanium	Nitrate	Tin
Soot (carbon)	Organic, biological		Lead
Water (except fog)	Magnesium		Vanadium
Lead	Aluminum		Copper
Ammonium ion	Silicon		Zinc
Hydrogen ion	Potassium		Antimony
Arsenic	Phosphate		Manganese
Selenium			



* THE FOUR AREAS NOTED INCLUDE PORTLAND, OREGON, LOS ANGELES, ST. LOUIS, AND WASHINGTON, D. C.

** 14 CITY STUDY (RECORD et al, 1976)

Figure 4-4. Results of microscopy/chemical analyses of urban particles. The bar chart is based on a large number of microscopic and chemical analyses as interpreted by an analyst (Draftz, 1980). The range of estimated concentration is based on a more limited data set collected in four areas where comprehensive mass balance studies were conducted (NAS, 1980). These are presented as an example of diversity of composition and size ranges. More graphic illustrations of the variations of particle composition and source types with time and geographic location can be found by examining Figures 5-47 to 5-49 of the criteria document.

substances in the fine mode, in rough order of mass, are water, ammonium sulfate and sulfuric acid, carbonaceous material (carbon and a variety of condensed organics), lead, nitrates, and small amounts of trace elements. These may take the form of mixed salt droplets, and a variety of heterogeneous particles with adsorbed gases, organics, or trace element as surface coatings. Sulfates and some other aerosols are hygroscopic (absorb water), growing to several times their original size (and mass) at higher relative humidities. This growth has important implications for deposition in the respiratory tract and for effects on visibility and climate.

The coarse fraction is more variable and less well characterized, containing a number of crustal and industrial minerals such as quartz, clays, limestone, and asbestos, rubber tire particles, as well as organics, nitrates and both road and sea salt. Salt is also hygroscopic. Among the trace substances that may be associated with coarse particles are pesticides, biological toxins, and soil dwelling microbes (Fraser and McDade, 1979).

2. Exceptions

The bimodal model of particulate matter is an extremely useful tool in describing typical aerosol distributions and elucidating formation and removal mechanisms and source contributions. This model also may be useful in considering welfare effects of particulate matter such as visibility or dustfall where a single mode clearly dominates. In evaluating the health effects of particles, however, conditions under which the distinction between modes becomes blurred may assume some importance.

Table 4-2 and Figure 4-4 list a number of important substances that, depending on location, apparently may be readily found in both modes. These substances include fly ash, coal fragments, minerals, nitrates, and trace elements such as vanadium. The relative contribution varies substantially. Under strong source or meteorological conditions that produce very high coarse or fine particle levels, the overlap between modes can increase substantially, thus making the demarcation between the two more difficult to detect. See, for example, the Pomona data of Figure 4-2 or Run 14 of Figure 4-3. In these cases, substantial fine or coarse particle mass may occur in the 2-5 μm range, which coincides with a maximum in deep lung deposition efficiency (CD, Figure 11-14). These extreme conditions of high concentration therefore may often be of more interest for primary standard setting than the more typical distributions encountered with lower levels.

3. Exposures

As a basis for comparison, Tables 4-3a and b provide estimates of typical ranges of TSP, dichotomous total (labeled Inhalable Particles or IP_{15}, <15 μm), and fine particles (<2.5 μm) as measured in three size specific particle sampling networks* for varying periods during 1977-81 (Pace, 1981). Analysis of limited temporal trends indicate that maximum seasonal averages for all three particle indicators occur in the summertime, particularly in the East (Pace et al., 1981). Overall ratios of IP_{15}/TSP in the most recent urban data remain fairly close to that reported by Pace (1980) for earlier data, which was 0.65 ± 0.12. Interpolation of TSP, IP, and FP results and examination of expected particle size distributions (Watson et al., 1981) suggest that the ratio of particle mass <10 μm (PM_{10}) to TSP in most areas would lie between about 0.5 to 0.6. Data on composition of these particles are not yet available.

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*The data used from the three networks include: EPA's Inhalable Particulate Network, 35 urban sites, April 1980-March 1981; EPA's Western Fine Particle Data Based, 40 non-urban sites, Summer 1980-Spring 1981; Electric Power Research Institute SURE data base, 9 rural sites, 15 months, 1977-1978 (Pace 1981).

TABLE 4-3. CHARACTERIZATION OF PARTICULATE MATTER CONCENTRATIONS FROM SIZE SPECIFIC NETWORKS, 1977-81 (Pace,1981)

a) LONG-TERM (6-12 MONTHS) AVERAGE	TSP	IP_{15}	FP
EASTERN LOCATIONS			
Undisturbed	30-40	25-35	15-20
Downtown	60-90	40-50	20-30
Industrial	60-110	45-70	25-45
ARID WESTERN LOCATIONS			
Undisturbed	15-20	10-15	3-5
Downtown	75-130	40-70	15-25
WEST COAST			
Los Angeles Area	90-180	50-110	30-40
Pacific Northwest	45-95	20-65	15-25

b) TYPICAL 24-HOUR MAXIMA*	TSP	IP_{15}	FP
EASTERN LOCATIONS			
Undisturbed	60-100	50-100	30-80
Downtown	90-210	75-140	40-90
Industrial	150-360	100-250	50-180
ARID WESTERN LOCATIONS			
Undisturbed	50-100	25-40	10-15
Downtown	125-310	70-180	45-70
WEST COAST			
Los Angeles Area	170-460	150-200	100-110
Pacific Northwest	115-310	50-190	45-90

*60 Samples/Year

Given reduced outdoor particle loadings and the trend toward year-round heating/cooling cycles, the relative contribution of indoor pollution to total particle exposures has clearly increased in the U.S. Indoor particle levels can be higher than outdoors because of smoking and other activities. For homes of non-smokers homes, the mass of smaller particles (e.g., sulfates) tends to be similar indoors and outdoors, but larger outdoor particles (as indicated by TSP) apparently do not completely penetrate (CD, p. 5-121 to 5-131). The size and compositional differences between indoor and outdoor particles, as well as routine daily activities, complicate the assessment of personal exposure and confound appraisal of both health and welfare (e.g., soiling) effects of particulate matter.

4. Comparison with Historical Composition

Although current particle levels are clearly lower than those reported in the historical London data, it is more difficult to assess differences in composition and comparisons are limited to sulfuric acid and primary carbonaceous material. Near major U.S. sulfur oxide sources, short-term peaks of 1 ppm SO_2 might be accompanied by primary sulfuric acid concentrations on the order of 50 $\mu g/m^3$*. Episodic levels of sulfuric acid equivalents in the range of 15-20 $\mu g/m^3$ may occur over large scale regions during the summer, accompanied by elevated oxidant levels (Lioy et al., 1980). These levels are an order of magnitude lower than the highest peaks reported during fall and winter British episodes but represent a greater proportion of particulate matter mass and may occur over far larger regions. The frequency of occurrence of such peaks, particularly in urban areas or during nighttime inversions where substantial ammonia neutralization may occur, is not well characterized. The relative proportion or primary organic and carbon particles probably is lower than that of London particles. This could change with increased use of residential wood or coal and passenger diesels. Secondary organic particle (e.g., oxygenated organics) levels probably are higher in the U.S. Peak levels of extractable organic particles can comprise as much as 50% or more of TSP in Southern California. Thus, the total extractable or volatizable organic fraction of particulate loading (as percent of TSP) in some U.S. cities can be equivalent to that of London (see Figure 4-4), but components within the organic fraction probably differ. Although it is reasonable to expect substantially higher quantities of fine particles during London episodes, reliable comparisons of other particle components (e.g., coarse carbon, dust) or of relative particle size distributions is not possible.

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*Assuming 1-2% of emitted SO_x is primary sulfuric acid. In this context "primary" refers to pollutants directly emitted from sources, while "secondary" pollutants are formed in the atmosphere from precursor materials as in the case of secondary particulate sulfates derived from oxidation of gaseous SO_2 .

V. CRITICAL ELEMENTS IN THE REVIEW OF THE PRIMARY STANDARD

A. Mechanisms

This section discusses the deposition and clearance of particles in the respiratory tract and the possible physiological and pathological responses to particulate substances. Implications of relevant deposition studies are assessed in light of current U.S. particle distributions and qualitative evidence from animal, controlled human, and epidemiological studies is used to identify potential mechanisms of toxicity. The major purposes of this discussion are to provide a basis for identifying the most appropriate indicator for those fractions of particulate matter of most concern to health and to examine plausible links between regional deposition of particles and observed responses in humans.

1. Particle Deposition and Clearance

a) Major Regional Divisions

An evaluation of the mechanisms by which inhaled particles ultimately may affect human health must recognize the importance of deposition and clearance in the respiratory tract. The major regions of the respiratory tract differ markedly in structure, size, function, and sensitivity or reactivity to deposited particles. They also have different mechanisms for particle elimination or clearance (Lippmann, 1977). As described by the criteria document (CD, Chapter 11), in discussing deposition and clearance, three major regions can be specified: extrathoracic, tracheobronchial, and pulmonary or alveolar. The nature and characteristic deposition and clearance mechanisms of these regions are outlined in Table 5-1.

The criteria document presents the range of available experimental deposition data for total and regional deposition in a series of figures

TABLE 5-1. MAJOR REGIONS OF THE RESPIRATORY TRACT (Lippmann, 1977; CD, Chapter 11).

Region	Description	Principal Size Range (s) Deposited	Major Mechanisms of Deposition	Major Mechanisms of Clearance	Normal Clearance Times for Insoluble Particles
Extrathoracic (ET)	The head and pharynx, down to and including the larynx. Subdivisions: Nose breathing Anterior nares Ciliated nasal passages Nasopharynx Mouth breathing Mouth, oropharynx	~1 - >100μm[a] hygroscopic aerosols >~0.3μm	Impaction	Mucociliary action to G.I. tract Sneezing, blowing, wiping to exterior Dissolution to bloodstream or mucous	minutes longer(nares)
Thoracic (TB+AL)					
Tracheobronchial (TB)	Ciliated conducting airways from the tracheal to the terminal ciliated bronchioles. (~ 16 generations).	~0.2 - 15μm[b]	Impaction Sedimentation Interception (Fibers)	Mucociliary action, coughing to G.I. Tract Dissolution to bloodstream or mucous	hours
Alveolar (AL) (Pulmonary)	Gas exchange region including unciliated airways and alveoli. Subdivisions: Respiratory bronchioles, alveolar ducts, alveolar sacs, atria, and alveoli.	<10 μm[c]	Diffusion(<1 μm) Sedimentation Impaction (>2 μm)	Phagocytosis to TB region, lymphatic system Dissolution to bloodstream, other fluids	weeks-years

[a] "Inspirability" Extrapolation by ISO, 1981.

[b] Extrapolation of NYU data by Miller et al., 1979.

[c] Figures 11-8, 11-9 criteria document.

(CD, Figs. 11-3 to 11-9). For simplicity, only alveolar and an estimate of tracheobronchial deposition (mouth breathing) are reproduced in Figure 5-1. A variety of subject related and environmental factors can influence deposition and clearance of aerosols, including inhalation patterns (rate and route), airway dimensions, disease state, particle composition, and the presence of pollutant gases. Their impacts are briefly summarized in Appendix A.

b) Deposition and Clearance of the Ambient Particle Distribution

The available data on respiratory tract deposition as discussed in Appendix A can be used to provide a preliminary qualitative evaluation of deposition of typically observed ambient particle distributions. Based on the deposition curves for normal nasal breathing in Chapter 11 of the criteria document, over half of the total mass of a "typical" mass distribution (see Section IV-2) would be deposited in the extrathoracic region, most of this being coarse particles. Clearance of most of this material to the esophagus would occur within minutes. Up to about half of the hygroscopic fine mass (e.g., sulfates and nitrates that grow to 2-4 μm in the respiratory tract) also might be deposited and dissolve in this same region. Smaller fractions (5-25%) of the hygroscopic and non-hygroscopic fine particles (mostly <1 μm) would be deposited in the tracheobronchial and alveolar regions, respectively. A similar fraction of coarse particles (size range 2.5 - 8 μm) will be deposited in these thoracic regions. Clearance of hygroscopic material by dissolution and reaction would be relatively rapid in both regions. Clearance of insoluble coarse mode substances would increase from less than an hour for the larger particles deposited in the upper portion of the tracheobronchial region to as much as a day for the more distally deposited. Insoluble fine and coarse particles deposited in

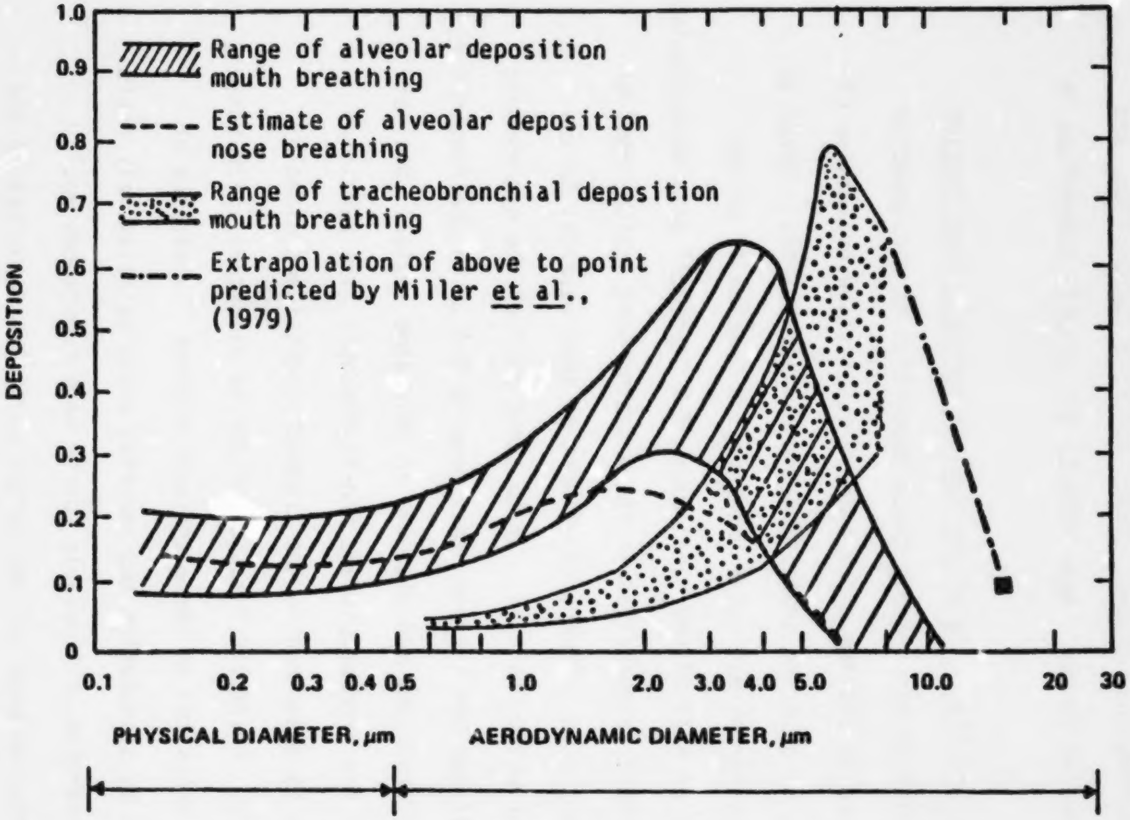

Figure 5-1. Regional deposition of monodisperse aerosols by indicated particle diameter for mouth breathing (alveolar, tracheobronchial) and nose breathing (alveolar). The alveolar band indicates the range of results found by different investigators using different subjects and flow parameters for alveolar deposition following mouth breathing. Variability is also expected following nasal inhalation. The tracheobronchial band indicates intersubject variability in deposition over the range of sizes as measured by Chan and Lippmann (1980). Deposition is expressed as fraction of particles entering the mouth (or nose) (CD, Figure 11-14). Also shown is an extrapolation of the upper bound of the TB curve to the point predicted by Miller et al. (1979). The extrapolation illustrates the likely shape of the curve in this size range but is uncertain.

the alveolar region would be expected to have lifetimes on the order of weeks to months or longer.

With mouth only breathing, the regional deposition pattern changes markedly, with extrathoracic deposition reduced and thoracic deposition enhanced. Extrathoracic deposition, although reduced, still would be dominated by coarse mode aerosols and contain little fine mode contribution. Both tracheobronchial and alveolar deposition of coarse mode and hygroscopic fine mode aerosols would be substantially increased (Figure 5-1). The higher endogenous ammonia in the mouth may, however, reduce the net deposition of acid aerosols (Larson _et_ _al_., 1977). Remaining fine particle deposition efficiency would be little changed over nasal breathing (<20%). .

In essence, regional deposition of ambient particles in the respiratory tract does not occur at divisions clearly corresponding to the atmospheric aerosol distributions. Coarse mode and hygroscopic fine mode particles are deposited in all three regions. A fraction (5-25%) of the remaining fine mode particles (e.g., organics and carbon not associated with hygroscopic material) is deposited in the tracheobronchial and alveolar regions. Little particle mass in excess of 15 μm is deposited in the thoracic region, and little mass greater than 10 μm is deposited in the alveolar region.

The above discussion represents a qualitative assessment of regional deposition of a "typical" ambient distribution for normal adults and breathing rates. Because atmospheric distributions and individual deposition characteristics, as well as other factors, vary widely, the above may not be representative of potentially important exposure/deposition cases. Considerations include:

1) Regional deposition data for mouth breathing tend to overestimate resting thoracic deposition for oronasal breathing and are strictly applicable

to those individuals who breathe only through the mouth. Nevertheless, a number of disease states and other conditions temporarily or permanently result in mouth only breathing. During exercise, even oronasal breathing can permit flow through the mouth comparable to or greater than used in mouth only studies (CD, p. 11-11). Moreover, the results of Hounam et al. (1969) and Andersen et al. (1979) suggest conditions or subject-related variability causing substantially lower nasal removal efficiency for coarse particles (in the 2.5 to over 12 μm range) than the ICRP model (CD, Figure 11-5) suggests. This would result in enhanced thoracic deposition for coarse particles during nasal breathing.

2) The peak in alveolar deposition efficiency for mouth and nose breathing (Figure 5-1) tends to occur at or near the normal minimum in the bimodal distribution (2-4 μm MMAD). The data of figures 4-2 and 4-3 suggest, however, that near sources or in other polluted conditions, substantial increases can occur in the coarse or fine mode contribution to this most efficiently deposited size range. Moreover, near sources a transient nuclei mass mode may be present (Figure 4-2). Theoretical models (Yu, 1978) and limited experimental results (CD, Figure 11-4) suggest that nuclei mode particles will be more efficiently deposited than accumulation mode aerosols.

3) As discussed in Appendix A, the deposition of both coarse and fine particles in the tracheobronchial region can be increased over "normal" ranges for: a) increased breathing rates during exercise; b) mouth breathing; c) cigarette smokers; d) bronchitics; and e) asthmatics. Although it reduces subsequent alveolar deposition, enhanced tracheobronchial deposition may not be protective, particularly for disease states such as bronchitis (Chan and Lippmann, 1980).

4) Regional mass deposition data do not provide insights into localized "hot spots". Significantly higher mass/lung surface ratios can occur in

the extrathoracic and tracheobronchial regions as compared to the alveolar region. As noted in Appendix A, enhanced deposition in the carina and other airway bifurcations should occur for both coarse and fine particles (Bell and Friedlander, 1973). Natural or deposition related slowing in clearance may occur in these areas (NAS, 1977a). Particles with surface coatings of toxic elements, organics, allergens, or gases (LaBelle et al., 1955; Natusch and Wallace, 1974) may result in greater effects than that suggested by total mass deposited because of initial localized surface reaction with tissue or macrophages (Camner et al., 1974a).

5) Although probability of deposition of particles larger than 10 μm in the alveolar region is low, small numbers of such particles have been found in human lungs (Michel et al., 1977). Some evidence suggests that those large insoluble coarse substances that do penetrate may be cleared at a much slower rate. Animal tests indicate that 15 μm particles instilled in this region clear much more slowly than smaller particles of the same composition (Snipes and Clem, 1981).

2. Mechanisms of Toxicity

Upon deposition, particles may produce physiological and ultimately pathological effects by a variety of mechanisms. Although the respiratory tract is equipped to remove inhaled foreign materials, if particle exposures are sufficiently high or if clearance mechanisms are impaired, accumulation of particles may reach levels that produce responses. Variations in subject related characteristics and aerosol physical and chemical composition play important roles in the nature of these responses. For purposes of discussion, the major mechanisms by which particles potentially may produce effects can be categorized as follows:

1) Chemical or mechanical irritation of tissues or nerve receptors at the site of deposition. This may result in immediate functional changes or cumulative insult. Substances as diverse as fine acid aerosols, insoluble coarse dusts, and SO_2/aerosol combinations can, in varying degrees, produce these responses in animals and humans.
2) Alteration of host defense systems, particularly clearance mechanisms. This may result in increased susceptibility to infection and potentiate development of chronic lung disease. Acid aerosols and, to a lesser extent, insoluble dusts might affect clearance rates by altering deposition patterns, increasing mucous secretion, or affecting the physiochemical properties of the mucous. Various particles may also damage alveolar macrophages.
3) Direct or indirect damage leading to morphological changes. This may lead to compromised lung function, potentiating development of or aggravating existing disease. Acid aerosols may produce direct tissue damage; certain coarse dusts, notably silica, may act indirectly through macrophage damage.
4) Systemic toxicity. Particularly toxic particles may be transported to and produce effects in other areas of the body. Examples include trace elements such as lead and arsenic or carcinogens such as polycyclic organics.

The extent to which these mechanisms may be operative depends strongly on the region of deposition, concentration, and relative chemical/toxicological activity of the aerosol, and the time the material remains in contact with respiratory surfaces. Pharmacological principles would indicate that response should be related to the dose delivered to the target area and hence be related to mass. In some cases, however, the effective dose may be related to other moments of the particle distribution, particularly surface area (NAS, 1977a; Natusch and Wallace, 1974). Particle deposition efficiency and surface area are inversely related in some regions of the respiratory tract, which may explain some apparently paradoxical animal and human study results. For example, depending on concentration, smaller particle sizes may result in either increased or decreased flow resistances in guinea pigs as compared to larger particle sizes (Amdur, 1958).

Table 5-2 presents a summary of potential mechanisms that might account for some of the responses and associations observed in toxicological and epidemiological studies of particulate matter. Potential mechanisms, consequences, and responses to particle deposition are organized according to the major regions of the respiratory tract. This summary is for qualitative purposes only; many of the mechanistic studies cited involve exposures significantly higher than those encountered under ambient conditions. The extent to which any one of these mechanisms explain observed epidemiological results or unexamined potential effects from ambient air exposure is speculative, but does serve to suggest which regions of the respiratory system are at greatest risk, based on current understanding. The following subsections briefly discuss some of the listed potential mechanisms following deposition by region. Discussion of studies relating to effects of concern is presented in Section V.B. and Appendix B.

a) Extrathoracic Deposition

The major ambient materials deposited in the extrathoracic region are coarse mode particles, with some contribution from hygroscopic fine particles. This region appears to be significantly less responsive to pollutant irritation than the tracheobronchial region (Nadel, 1973; Widdicombe

TABLE 5-2. POSSIBLE RESPONSES TO PARTICLE DEPOSITION IN THE RESPIRATORY TRACT

Principal Region of Deposition	Potential Mechanisms	Potential Consequences/Observations
Extrathoracic (ET)	Chemical and mechanical irritation/stimulation of receptors by deposited material	Symptomatic Effects: -Dryness in nose, mouth, and throat (polymerized dust containing carbon black, Andersen et al., 1979) -Sneezing, rhinitis (pollen), (Michel et al., 1977)
	Slowed, stopped mucociliary clearance by wood dust (Schlesinger and Lippmann, 1978)	Nasal cancer (wood workers, NAS, 1977a, p. 158)
	Enhanced deposition at larynx (Schlesinger and Lippmann, 1976)	Laryngeal cancer (cigarette smoke, NAS, 1977a, p. 158)
Tracheobronchial (TB)	Chemical and mechanical irritation/stimulation resulting in bronchoconstriction (Widdicombe et al., 1962; Nadel, 1973) by: ."Inert" dusts (granulated charcoal, coal dust, carbon dust, calcium carbonate, carbon impregnated plastic, iron hydroxide; Widdicombe et al., 1962; Dubois and Dautrebande, 1958; Andersen et al., 1979; Constantine et al., 1959) .Resuspended urban dust (crustal materials, sulfates, volatiles; Toyama, 1964) .Coarse organic dusts, aeroallergens (grain dusts, pollens, mold, etc. e.g.; Dosman, 1980) .Fine acid aerosols (sulfuric acid, ammonium bisulfate; Utell et al., 1981) .Community air pollution with moderately high PM (Lebowitz et al., 1974) .Fine particles in combination with pollutant gases (SO_2; Koenig et al., 1981; McJilton et al., 1976)	Reduced respiratory function Enhanced breathing difficulties or other acute aggravation of heart and lung disease, including: -Asthma (Smith and Paulus, 1971) -Bronchitis (Lawther et al., 1970) -Emphysema and cardiovascular disease (Martin and Bradley, 1960) -Influenza (sodium nitrate, Utell et al., 1980) Enhanced deposition of fine and coarse particles (Albert et al., 1973)
	Enhanced sensitivity to subsequent bronchoconstrictive agents by sulfuric acid (Utell et al., 1981)	As above
	Altered clearance rates, increased mucous production by deposited material (cigarette smoke, sulfuric acid, dusts) (Lippmann et al., 1981; Camner et al., 1973)	Possible promotion of bronchitis by repeated exposure to sulfuric acid (Lippmann et al., 1981) -Increased bronchitis prevalence in people exposed to community air pollution (Holland et al., 1969; Holland and Reid, 1965) -Increased bronchitis prevalence in workers exposed to coal dust, other dusts (Morgan, 1978)
	Direct damage to tissues by acid aerosols	Bronchial lesions (Alarie et al., 1975)
	Increased deposition at bronchial bifurcations (Bell and Friedlander, 1973), slower clearance (Hilding, 1957)	Sites of enhanced deposition are most frequent sites of bronchogenic carcinoma (Schlesinger and Lippmann, 1978)
	Interaction of carcinogens and ambient particles	Intratracheal BaP tumor production potentiated by carbon, iron ore, asbestos (Pylev and Shabad, 1973; Stenbäck et al., 1976; Saffioti et al., 1968) Increased lung cancer among urban vs. rural smokers (Higgins, 1976)

TABLE 5-2. POSSIBLE RESPONSES TO PARTICLE DEPOSITION IN THE RESPIRATORY TRACT (CONTINUED)

Principal Region of Deposition	Potential Mechanisms	Potential Consequences/Observations
Pulmonary (Alveolar, AL)	Decreased oxygen transport and probable increased resistance of blood flow through pulmonary capillaries	Aggravation of cardio-pulmonary disease associated with -London air pollution (Ministry of Health, 1954; Martin, 1964)
	Death of macrophages resulting in release of proteolytic enzymes that damage alveolar tissues, by silica, other coarse dusts (Ziskind et al., 1976)	Pneumoconiosis -Sand in desert dwellers (Bar-Ziv and Goldberg, 1974) -Agricultural dust in farm workers (Sherwin et al., 1979 -Dust in zoo animals (Brambilla et al., 1979) -Silica in granite workers (Craighead and Vallyathan, 1980) -Slate dust in miners/quarrymen (Glover et al., 1980) -Coal dust in miners (Morgan, 1978) -Fibrosis followed exposure to chrysotile asbestos (NIOSH, 1976)
	Damage to macrophages, other host defense mechanisms by surface coating of toxic materials (Camner et al., 1974a; Aranyi et al., 1979) Damage to tissues by acid aerosols	Increased susceptibility to infection -Increased mortality in infected mice exposed to Cd, Ni, Mn aerosols (Gardner 1981; Graham et al., 1978; Adkins et al., 1979, 1980) -Loss of alveolar surface area ("emphysema") in dogs exposed to H_2SO_4 (Hyde et al., 1978)
	Combined effect of exposure and slow clearance of particles (Pratt and Kilburn, 1971)	Accumulation of pigment in lungs from inhaled particulate matter (Pratt and Kilburn, 1971; Sweet et al., 1978) Possible role of community air pollution in emphysema (Ishikawa et al., 1969)
Thoracic, not specific to TB, AL	Possible effects on host mechanisms (clearance, immunology) promoting infection	Increased lower respiratory tract infection in children with increased BS+SO_x exposure (Douglas and Waller, 1966; Lunn et al., 1967) Increased influenza rates ("Dust"; Kalpazanov et al., 1976)
	Successive measurements of respiratory function suggest damage to the lung during childhood may be produced by infection (Speizer et al., 1980)	Persistent changes in airways in those children with higher infection rates previously exposed to particulate matter (Colley et al., 1973; Kiernan et al., 1976)
	Absorption of systemic toxicants (e.g. pesticides, trace elements, carcinogens) resulting in extra-respiratory effects. Absorption efficiency greatest for alveolar deposition	Hematological and nervous system effects of lead (EPA, 1977) Gastro-intestinal cancer (Winklestein and Kantor, 1967)

et al., 1962). Exposures of normal subjects to high levels of coarse "inert" dust* (carbon impregnated plastic) resulted in a mild increase in symptoms, especially dryness of the nose and throat, but no effect on mucociliary clearance (Andersen et al., 1979). Studies of nasal exposure of normal subjects to fine sulfuric acid aerosol for 1 hour at levels of 110-980 $\mu g/m^3$ reported no extrathoracic related symptoms (Leikauf et al., 1981), but particle clearance in this region was not measured. Oral exposure to sulfuric acid (220-940 $\mu g/m^3$) produced throat irritation that affected greater numbers of subjects with higher concentrations (Horvath et al., 1981). Deposition of pollens or other allergens may result in rhinitis, the major nasal response to irritation. The findings of slowed nasal mucociliary clearance from exposure to wood dust (Schlesinger and Lippmann, 1978) and SO_2 (Andersen et al., 1974) suggest that additional study is needed on the effects of aerosol exposure on nasal mucociliary clearance, especially in view of the findings of nasal cancer in wood workers. Available information provides no indication of substantial effects on clearance at typical ambient levels.

b) Tracheobronchial Deposition

The tracheobronchial region is exposed to a mixture of fine (particularly hygroscopic fine) and coarse particles. Among the major effects possibly associated with deposition of particles in this region are bronchoconstriction, altered clearance, aggravation and promotion of chronic respiratory diseases (e.g. asthma, bronchitis) and cancer.

Bronchoconstriction is one of the most common responses associated with particle exposure and has been reported following short-term exposure to high concentrations of a variety of "inert" dusts, and acidic and alkaline aerosols of varying particle sizes (<1 to over 10 μm). Current evidence suggests that bronchoconstriction produced by acute exposures is probably a reflex reaction resulting from chemical and/or mechanical stimulation of irritant neural receptors (Widdicombe *et al.*, 1962). Although dose/response characteristics vary among substances, both dusts and chemically active substances appear to induce bronchoconstriction in a similar manner because bronchoconstriction can be blocked by the administration of drugs or surgical removal of nerves (Nadel *et al.*, 1967; Widdicombe *et al.*, 1962; Toyama, 1964).

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*This synthetic particle as well as a number of insoluble mineral dusts are often termed "inert" because of their general chemical and biological stability. Because such materials can produce biological responses, however, quotation marks are used.

Because deposition is elevated at, and epithelial nerve endings tend to be concentrated near, airway bifurcations, deposition at bifurcations may well have a major influence on pulmonary mechanical changes induced by either mechanical or chemical stimulation of receptors (Nadel *et al.*, 1967). The nerve endings of the more central airways are especially receptive to mechanical stimulation that lead to reflex coughing and bronchoconstriction.

Alterations of lung function in this region may be, in part, related to effects observed in epidemiological studies of particle exposure, such as the aggravation of chronic respiratory disease, including asthma, bronchitis and emphysema. The airways of asthmatics are hyperreactive to a variety of irritants (Boushey *et al.*, 1980). Individuals with chronic obstructive lung disease may not be hyperreactive, but typically have abnormal lung function. Further depressions in lung functions by particle exposures that may be of no consequence to normal individuals could be incapacitating for individuals with asthma, or even life threatening for individuals in the later stages of chronic lung disease suffering some degree of oxygen deficiency (hypoxia) (Norris and Bishop, 1966). Moreover, as indicated earlier, individuals with asthma and bronchitis may have significantly higher deposition rates in the tracheobronchial region (Albert *et al.*, 1973). This enhanced particle deposition may trigger further bronchoconstriction and further increase deposition above that in the initial exposure condition.

Another carefully studied biological endpoint in the tracheobronchial region is mucociliary clearance. As with bronchoconstriction, pollutant effects on mucociliary transport involves mediation, in part, through the nervous system (Camner _et_ _al_., 1974b; Camner _et_ _al_., 1976). As indicated in Table 5-2, human studies have found that mucociliary clearance is affected by fine sulfuric acid, high levels of coarse carbon dust, and cigarette smoke. Substances that affect clearance do not always cause bronchoconstriction at similar levels (Lippmann _et_ _al_., 1981).

Repeated exposures to substances that alter mucociliary clearance may potentiate development of chronic obstructive pulmonary disease. Lippmann _et_ _al_. (1981) proposes the following sequence of events in producing chronic bronchitis: Acute high level exposures to particles increase mucus production and mucociliary transport. Continuation of these exposures leads to bronchial mucous gland hypertrophy and goblet cell hyperplasia. Mucus transport in small airways might be normal or increased during this stage. As gland hypertrophy continues, the mucociliary transport system becomes inadequate in removing mucous secretions. This leads to chronic cough, accumulation of secretions, and further susceptibility to inhaled particles, noxious gases, and pathogenic organisms.

Following this sequence of events, particulate exposure may be: 1) causal factors in chronic bronchitis; 2) predisposing factors to acute bacterial and viral bronchitis, especially among children and cigarette smokers; and 3) aggravating factors for acute bronchial asthma and the terminal stages of oxygen deficiency associated with chronic bronchitis and/or emphysema and its characteristic form of heart failure (cor pulmonale) (NAS, 1977a). Increased prevalence of bronchitis has been observed in cigarette smokers and people exposed to high community particle and sulfur oxide pollution (Holland and Reid, 1965) or various occupationally derived mineral dusts (Morgan, 1978).

In vitro studies suggest that some particles deposited in the tracheobronchial region may become attached to the mucosal surfaces of the airways (Mossman *et al.*, 1978). Larger particles (10-15 μm) can be lodged on tracheal surfaces despite continued mucociliary action; these particles have been associated with the *in vitro* sloughing of cells shortly thereafter. Smaller particles (0.5-1 μm) may be transported to the submucosa where they can be taken up by mesenchymal cells of the trachea (Mossman *et al.*, 1978). These findings indicate that not all insoluble particles are necessarily rapidly cleared from the tracheobronchial region by mucociliary action.

As noted above, the bifurcations of the larger airways appear to be especially vulnerable to the accumulation of particles. Increased deposition and potentially resulting slower clearance (Hilding, 1957) in these sites are correlated with major locations of lung tumors (Schlesinger and Lippmann, 1978).

c) Pulmonary (Alveolar) Deposition

Particle deposition within the alveolar region of the lungs is essentially limited to particles less than 10 μm. Several important characteristics in the alveolar region affect response to particle inhalation. As indicated earlier, clearance from the alveolar region of the lungs is much slower than from the tracheobronchial region. The alveolar region is the site of oxygen uptake and of various non-respiratory functions of the lungs that may be affected by pollutant exposures.

Many victims of the London air pollution episodes were patients suffering from cardiopulmonary disease and were thought to have emphysema and bronchitis. Such diseases normally reduce the ability of the lung to transfer oxygen to the blood. It is hypothesized that the disturbance of normal ventilation and perfusion in the lung by acute pollution exposure may shunt air from some alveoli so that a sudden lack of available oxygen may be experienced. A reflex constriction of blood vessels supplying part of the lungs may follow, resulting in increased pulmonary arterial pressure. Although this added load (associated with pollution exposure) should be tolerable in normal individuals, the added stress and chain of events could possibly lead to fatal or irreversible damage in individuals compromised with cardiopulmonary disease.

The alveolar region of the lung is the site of several respiratory diseases that are associated with inhalation of particles. Long term exposures to high levels of acid aerosols, as well as some other pollutants, have produced conditions resembling emphysema in controlled animal exposure studies (Hyde et al., 1978). Various animal, occupational, and community exposures to insoluble particles, including coal dusts, silica, asbestos, and ambient dust may result in accumulation of material in lungs, and possibly through damage to macrophages, are associated with inflammation, fibrosis and other conditions (Brambilla et al., 1979; Sherwin et al., 1979; Ziskind et al., 1976). This "dust lung disease" or pneumoconiosis may vary in severity and pathological condition.

d) Thoracic Deposition, (Not Necessarily Specific to TB, AL Regions)

Infection may be initiated in any area and spread to other regions. Exposure to specific particle components has been shown to affect clearance (Leikauf et al., 1981) and the immune system (Zarkower, 1972), both of which are considered to be important host defense mechanisms against infection.

Observational studies show that exposure to particle/sulfur oxide pollution is related to increased prevalence rates of infection among children (Lunn et al., 1967). Observational studies also indicate that when children experience a history of greater respiratory illness (associated with pollutant exposure), these illnesses may produce small decrements in pulmonary mechanics. Speizer et al. (1980) speculate that the lungs of those children affected by respiratory diseases associated with pollutant exposure ultimately may not develop full potential capacity as adults. Long term British studies of individuals born in 1945 (Douglas and Waller, 1966; Colley et al. 1973; Kiernan et al., 1976) appear to support this possible mechanism in that a history of childhood respiratory disease is associated with persistent changes in airways as indicated by increased prevalence of respiratory illness in later years.

Systemic toxicity may result from deposition in any region. Some constituents of particulate matter produce systemic toxicity when they enter the blood and are transported to extra-respiratory sites. Systemic toxicity has been shown following inhalation of several metals including lead (EPA, 1977), mercury (Hammond and Beliles, 1980), arsenic (NAS, 1977b), cadmium (Hammond and Beliles, 1980), and some organic substances (NAS, 1972). Clearance of inhaled carcinogenic particles to the gastrointestinal tract might account for the observed association of elevated gastrointestinal cancer with high particle levels (Winklestein and Kantor, 1967). The observed association is, however, questionable (CD, p. 14-83).

B. Effects of Concern

This section identifies and describes the principal effects of concern associated with exposure to particulate matter. Evidence for such associations drawn from animal toxicology, controlled human exposures, and community epidemiological studies is discussed and evaluated in Appendix B. Based on these data, the following effects areas appear to be of most interest:

1) Respiratory Mechanics and Symptoms;
2) Aggravation of Existing Respiratory and Cardiovascular Disease;
3) Clearance and other Host Defense Mechanisms:
4) Morphological Alterations;
5) Carcinogenesis; and
6) Mortality.

The major implications of the literature evaluated in Appendix B relating to each of these effects areas are briefly summarized below.

1. Respiratory Mechanics and Symptoms

Effects on respiratory mechanics can range from mild transient changes of little direct health consequence to incapacitating impairment of breathing. Symptomatic effects also vary in severity, but at minimum indicate a biological response. Key conclusions with respect to the effects of particles include:

1) A number of long-term observational studies (Table B-3) have found that populations living in areas with high particulate pollution (usually with high SO_2) tend to have a higher prevalance of respiratory symptoms and lower lung function capability compared with other groups in areas with lower pollution levels. When differences in particle concentrations are small, effects are difficult to detect.

2) Only two community studies (Lebowitz _et al_., 1974; Dockery _et al_., 1981) have reported lung function decrements (in children and adolescents, some exercising) in association with short-term particle exposures. Occupational studies, controlled human expoures, and animal studies

discussed in Appendix B suggest that a variety of particles can, at high levels, produce relatively rapid functional changes.

3) Most short-term (10 minutes-4 hours) animal and human investigations of sulfuric acid and other sulfates have found no direct respiratory mechanical changes below 1000 $\mu g/m^3$, a level higher than those seen even in peak London episodes (Table B-2). Asthmatics show some enhanced response, but other sensitive groups and longer exposures (> 2-4 hours) have not been tested. Responses to sulfates in both asthmatics and normals generally decrease with lower particle acidity. Differences in subject sensitivity and health status may be in part responsible for some observations of respiratory mechanical changes at lower sulfuric acid levels in guinea pigs (100-1000 $\mu g/m^3$) and humans (350 $\mu g/m^3$). The importance of health status is illustrated by the reactivity of people with influenza to an apparently neutral nitrate salt (Utell _et al_., 1980), at high concentrations that produced no responses when the same subjects were healthy.

4) Short-term exposures (minutes - 4 hours) to various insoluble particles and resuspended dustfall at high levels (2 to > 50 mg/m^3) have produced variable degrees of bronchoconstriction and respiratory symptoms in normal healthy adults and in diseased subjects. In some of these studies, coarse dusts (2 to > 12 μm) are clearly implicated (McDermott, 1962; McKerrow, 1964; Andersen _et al_., 1979; Toyama, 1964). Although some negative findings are reported, the lower concentrations tested and found to produce responses in some subjects (2-10 mg/m^3) are in the range of peak 1-hour smoke levels reported in London

(10 mg/m^3), and within an order of magnitude of likely hourly peaks during less severe British and U.S. episodes.

5) Particles can enhance penetration and toxicity of soluble pollutant gases, notably, SO_2 (McJilton _et_ _al_., 1976; Amdur and Underhill, 1968). The nature of the interaction is a matter of some complexity and debate (NAS, 1978). The data suggest that higher particle solubility and surface area increase interaction potential.

2. Aggravation of Existing Respiratory and Cardiovascular Disease

Particle-induced brochoconstriction, mucous secretion, and other effects might aggravate those with respiratory and cardiovascular disease. A number of community observational studies (Table B-4) of episodic as well as more moderate peak exposures to particles (usually with SO_2) suggest that these exposures aggravate the conditions of cardiovascular patients and, individuals with bronchitis, emphysema, pneumonia, asthma, and influenza.

3. Alterations in Host Defense Mechanisms

The major mechanisms discussed include clearance of particles and other foreign matter from the respiratory tract and other respiratory system-related defenses against infectious agents. Important conclusions include:

1) Studies of animals (donkeys) and humans suggest that exposure to repeated short-term peaks of sulfuric acid (100 $\mu g/m^3$, 1 hour/day, 6 months) may produce long-term slowing of mucociliary clearance, and by analogy to cigarette smoke, might play a role in the etiology of chronic bronchitis (Lippmann _et_ _al_., 1981). These sulfuric acid

levels are roughly within an order of magnitude of possible repeated U.S. peaks, and within the range of maximal London episode peaks (Section IV).

2) Community epidemiological studies (Holland and Reid, 1965; Lambert and Reid, 1970) of particles and sulfur oxides and occupational studies of industrial bronchitis in workers exposed to high dust levels (Morgan, 1978) suggest that high long-term particle exposure is associated with an increase in the prevalence of bronchitis.

3) Animal exposure studies show that particles of various types may damage alveolar macrophages and affect immunology (CD, Section 12.3.4.2). Animal infectivity model results do not provide convincing evidence of enhanced mortality following administration of pathogens and exposure to particle components commonly found in the ambient air, with the possible exceptions of high levels of sulfuric acid and carbon or ozone exposure mixtures. The relevance of these results to human exposures is unclear.

4) Community studies (Table B-7) suggest increased infectious disease during pollution episodes and in children and adults living in areas of higher particle and SO_2 pollution. As noted in Table 5-2, such infections in children might have longer term consequences.

4. Morphological Damage

Long-term exposures to particles damage lung tissue, but the available evidence is mainly qualitative. Specifically:

1) Multi-year studies of dogs exposed to sulfuric acid/SO_2 mixtures (Table B-8) indicate damage to the tracheobronchial region and

alveolar lesions that are analogous to an incipient stage of emphysema. Damage apparently continued to increase following the termination of exposure. Several factors outlined in Appendix B complicate application of the results to real world human exposures.

2) 12-18 month exposures of monkeys and guinea pigs to various mixtures of sulfuric acid, SO_2, and fly ash suggest that most morphological damage reported was due to sulfuric acid perhaps in combination with ammonium sulfate salts (Table B-8). Although the fly ash particles were somewhat large for deep lung penetration in this small animal, some accumulation was noted, but without obvious effect. Post exposure examinations similar to those in the above dog studies were not performed.

Retrospective autopsy studies (Table B-9) of animals and humans exposed to various crustal dusts at near to slightly above ambient levels suggest these exposures result in silicate pneumoconiosis. Responses range from the build up of particles in macrophages with no clear clinical significance to possibly pathological fibrotic lesions. The studies suggest deposition of ambient coarse particles in the alveolar region is of some concern but do not provide a basis for quantitative health assessments.

5. Carcinogenesis

Cigarette smoking is generally considered to be the major determinant of lung cancer (Higgins, 1976; Doll, 1978). The high particulate air pollution of the 1940-60's contained carcinogens and may have potentiated or otherwise contributed to elevated cancer rates in urban areas. Based on filter chemical analyses, levels of some particulate carcinogens have declined since that time and hence carcinogenic risk from these substances presumably has been reduced. The available evidence does not unequivocally show that current particle exposures contribute to cancer. Neither does it disprove any effect. Because lung cancer is the leading cause of cancer-related deaths, the risk of a small effect could assume some importance. The presence of mutagens in organic particulate fractions from unidentified sources (Pitts et al., 1977) and potential interaction between these or inert particles and carcinogens from cigarettes or occupational exposures suggests some need for caution and further study.

6. Mortality

A number of epidemiological studies demonstrate an association between peak particle and SO_2 pollution and mortality in certain sensitive populations (Table B-10). While some analyses suggest that the observed pollution effects may be more attributable to particles, it is not clear that the available data permit such a determination.

Regression analyses of long term mortality and pollution data (Table B-11) have been used to suggest that sulfur-containing particulate matter at current ambient levels is associated with excess mortality. A number of inherent difficulties confound such studies to the extent that they do not provide reliable evidence on such effects of particles and sulfur oxides. They are not, however, inconsistent with the possibility of associations at levels below those observed in the more extreme episodes.

C. Sensitive Population Groups

This section identifies those groups most likely to be among the most sensitive to the effects of particulate matter, based principally on material presented in Sections A and B. Available estimates of the size of each of these groups in the U.S. population are also given.

TABLE 5-3. SENSITIVE POPULATION SUBGROUPS

Subgroup	Population Estimates	Rationale (or Criteria)	Observational/Associations Supporting Increased Sensitivity
Individuals with chronic obstructive pulmonary diseases . Bronchitis . Bronchiectasis . Emphysema	7,800,000 (DHEW, 1973)	-Mucus hypersecretion and blocked airways may predispose individuals to bronchospasm -Enlarged airspaces increase blood flow resistance through the pulmonary capillary network, increasing cardiac stress	Many of the deaths and illnesses during and after air pollution episodes were among people with pre-existing obstructive diseases (Ministry of Health, 1954; Martin, 1964; Lawther et al., 1970; Martin and Bradley, 1960)
Individuals with cardiovascular disease	16,100,000 (DOC, 1980)	-Enhanced sensitivity to difficulties in breathing	Many deaths and hospitalizations during pollution episodes among cardiovascular patients (Ministry of Health, U.K., 1954; Martin, 1964)
Individuals with influenza	Unknown	-Increased sensitivity of respiratory epithelium (Utell et al., 1980)	Influenza patients were more sensitive to $NaNO_3$ during their period of sickness (Utell et al., 1980). Highest mortality during influenza epidemic on days with highest PM. (Martin and Bradley, 1960).
Asthmatics	6,000,000 (DHEW, 1973)	-Hyperreactive airways (Boushey et al., 1980)	Sulfuric acid enhanced response to bronchoconstrictive agent in asthmatics, not in normals (Utell et al., 1981)
Elderly	24,658,000 >65 years old (DOC, 1980)	-Reduced lung elasticity (Cotes, 1979) -Immunologically deficient	-Many of the deaths and illnesses during air pollution episodes were among elderly (Ministry of Health, 1954; Martin and Bradley, 1960; Greenburg et al., 1962).
Children	46,300,000 >14 years old (DOC, 1980)	-Immunological immaturity implies diminished protection (Eisen, 1976) -Childhood respiratory infection might prevent the lungs from reaching their full size at maturity (Bouhuys, 1977; Speizer et al., 1980) -Children likely to spend a greater amount of time outdoors and to be more active. Probably higher ventilation rates and thus, increased inhalation of pollutants.	-Increased acute respiratory disease with high particles, SO_x (Lebowitz et al., 1972; Douglas and Waller, 1966) -Effects of acute respiratory disease acquired during childhood persisted until adolescence or young adulthood (Colley et al., 1973; Kiernan et al., 1976).
Smokers	50,000,000 (DHEW, 1977)	-Urban lung cancer in smokers greater (Doll, 1978) -Combinations of PM and carcinogens may enhance response -Increased tracheobronchial deposition (Albert et al., 1973)	-Frequency of respiratory symptoms and diseases greater in smokers exposed to same occupational or community pollution as non-smokers (Lambert and Reid, 1970; NIOSH, 1976).
Mouth or oronasal breathers	15% of population (Niinimaa et al., 1981; Saibene et al., 1978)	-Increased particle penetration (CD, p. 11-20)	

The presence of certain diseases appears to be an important factor in the predisposition of certain individuals to the harmful effects of particulate matter. The history of catastrophic pollution episodes (Firket, 1931; Schrenk et al., 1949; Ministry of Health, 1954) as well as the lesser episodes in London (Martin, 1964) has clearly indicated that chronically ill individuals, especially those with cardiovascular and respiratory diseases, as well as the very young and the very old, were more severely affected than other groups.

Data derived from lower levels of community exposures and from other sources suggest additional segments of the population that may be at higher than average risk. These include children, asthmatics, smokers, obligatory mouth breathers, and individuals with pneumoconiosis or influenza. Table 5-3 draws upon information from epidemiological, toxicological, human clinical and physiological research in summarizing the observations that have identified various subgroups and possible explanations for their sensitivity. Much individual variation exists among the sub-groups. For example, at any given level of particles, some exposed children may note only symptomatic irritation, while others may suffer deterioration of respiratory function. Moreover, in some respects, these groups can sometimes be at lower risk, as in the case of influenza patients confined to indoor environments with lower than ambient pollution levels.

Data from the U.S. National Health Safety Survey for 1970 indicates that chronic respiratory disease makes up ten percent of all conditions causing disability of one week or more. In 1970, there were about 6.5 million chronic bronchitics, 6.0 million asthmatics, 1.3 million individuals with emphysema and about 10 million adults with heart disease severe enough to limit activity (DHEW, 1973). These are rough estimates since some surveys have reported higher figures depending on age, sex, and the definition of disease that is used. Limited physiological studies suggest that about 15% of the population are habitual mouth or oronasal breathers (Saibene, et al., 1978; Niinimaa et al., 1981). Anyone may temporarily switch to mouth breathing during exercise, illness, and conversation.

Although there are about 50 million smokers, the number of people at a higher than expected risk because of smoking may also include children living with smokers, and ex-smokers (DHEW, 1977). In addition, some workers who are occupationally exposed to dusts might become more susceptible to community particulate pollution, even if they are not classified as having respiratory disease. The more sensitive individuals, however, often do not remain in such work environments (Morgan, 1978).

D. Concentration/Response Information

As outlined in Section B, reactions to particulate matter can be divided into acute and chronic effects in healthy individuals as well as those with existing pulmonary or cardiovascular diseases. Air pollution clearly has been associated with an increase in both mortality and morbidity during smog episodes, as well as impaired lung function and increased respiratory symptoms among individuals with long-term high-level exposures. The literature summarized in Appendix B shows that the existence of these general associations has been demonstrated in many countries and population groups, and is at least qualitatively supported by controlled laboratory exposures of animals and humans to various components of particulate matter.

Trying to assess the precise level of particulate pollution associated with observed effects on health, however, has many problems. Suspended particulate matter is not a uniquely defined entity. The comprehensive physical and chemical characteristics are not only hard to measure and relate to health effects, but vary with monitoring device, geography, and time. This increases the uncertainty of any extrapolations from one set of circumstances to another, and greatly limits the utility of laboratory studies of single substances for quantitative purposes.

Epidemiological studies can provide strong evidence for the existence of pollutant effects, but are more limited for identifying accurate effects levels for specific pollutants or pollutant classes. Among the more important limitations of epidemiology as discussed in Section 14.1.1 of the criteria document are: 1) inadequate and inconsistant measurement of the exposure burden of individuals; 2) variability in the measurement of health endpoints (e.g., lung function, hospital admissions, frequency of symptoms) and in the sensitivity of populations studied; 3) failure, especially in cross-sectional studies, to control fully for confounding, or covarying factors, such as cigarette smoking and socioeconomic status; 4) difficulty in distinguishing particles from other pollutants; and 5) inability to establish a causal relationship, or negate one, based only on statistical associations.

Recognizing these limitations, epidemiological studies must still form the principal basis for developing concentration response assessments for particulate pollution. The following review summarizes those studies cited by the criteria document as providing the most reliable quantitative information as well as other studies that provide reasonable evidence of exposure response relationships without allowing derivation of specific levels. A further assessment of these studies as applied to selecting alternative levels for air quality standards is presented in Section VI.

1. Acute Exposures

a) Mortality

During the historical London pollution episodes, significant increases in mortality occurred when daily averages of British smoke exceeded 1000 $\mu g/m^3$ together with daily average SO_2 levels in excess of 1000 $\mu g/m^3$ (Ministry of Health, 1954; Scott, 1963). Smaller increases in mortality also have been observed in episodes in New York (Glasser et al., 1967). The criteria document states that these studies "tentatively suggest that small increases in excess mortality may have occurred at simultaneous elevations of 1000 $\mu g/m^3$ SO_2 and PM above 5.0-8.0 CoH, but this is much less clearly established "(CD, p. 14-15). Most of these episode-related effects were among particularly vulnerable sections of the population, such as the elderly and those with cardiopulmonary disease.

Studies of pollution/mortality associations during less extreme conditions are more relevant for current purposes. Several studies, listed in Table 5-4, have examined minor or non-episodic variations in pollution. Martin and Bradley (1960) related daily mortality from all causes (and from bronchitis and pneumonia) to daily levels of smoke and SO_2 in London during the winter of 1958/59 which was an unusually foggy period. A considerable number of coincident peaks in pollution concentration and daily mortality were found. British smoke and SO_2 correlated highly with mortality from all causes. An influenza epidemic during the month of February may have confounded the results. The correlation between mortality and mean daily temperature was not significant and

TABLE 5-4. SUMMARY OF EPIDEMIOLOGICAL STUDIES PROVIDING MOST USEFUL CONCENTRATION/RESPONSE INFORMATION FOR ACUTE PARTICLE EXPOSURES

Observed Effects	Time	Population	Levels Where Effects Are Likely (CD, Table 14-7)		Comments	Study
			BS (as $\mu g/m^3$	SO_2 ($\mu g/m^3$)		
Clear increases in daily mortality	1958-60 winters	Metropolitan London	$\geq$ 1000	$\geq$ 1000	1958-59 winter episodes. Unusually foggy with peak pollution. (Lawther, 1963; Holland et al., 1979).	Martin and Bradley 1960 Martin, 1964
Likely increases in daily mortality	1958-60 winters 1958-72 winters	Metropolitan London	500-1000	500-1000	"Greatest certainty" of increases with BS, SO_2 > 750 $\mu g/m^3$, but indications of small increases with BS < 500 $\mu g/m^3$, possibly as low as 150-200 $\mu g/m^3$ (CD, Table 14-7). Significant correlation most consistent for smoke.	Martin and Bradley 1960 Martin, 1964 Ware et al., 1981 Mazumdar et al., 1981
Daily worsening of health status in bronchitics	1954-68 winters	180, and later ≈ 1000 chronic bronchitic patients, London	250-500	500-600	Well designed. Peak values above daily average may have been important. Minimum levels for any "significant" response were 250 $\mu g/m^3$ BS, 500 $\mu g/m^3$ SO_2; significant associations among sensitive subgroup at lower levels. (Lawther et al., 1970).	Lawther et al., 1958; 1970

only marginally significant for humidity. Visibility, which is influenced both by fog and particulate pollution, was also highly correlated with mortality.

Martin (1964) analyzed the winter of 1959-60 in which there were fewer incidents of high pollution and still found significant positive correlations between mortality and pollution. Both studies relied on measurements taken from a seven station monitoring network in central London that, according to the criteria document, was a good index of city-wide concentrations. The errors for individual measurements are difficult to calculate. Ware et al. (1981) re-examined the data from both winters after excluding the month of the influenza epidemic and provided evidence that positive deviations in mortality occurred with BS levels greater than 500 $\mu g/m^3$ with increasingly negative deviations below that level. The criteria document points out that it is unclear at what level significant excess mortality occurred, but concludes that notable increases in excess mortality occurred in the range of 500-1000 $\mu g/m^3$ BS and SO_2 and were most likely when both pollutants exceeded 750 $\mu g/m^3$; small increases in mortality may also have occurred at levels below 500 $\mu g/m^3$ of either pollutant (CD, Table 14-7).

The Ware et al. (1981) and some other analyses (CD, Appendix 14-D) omit data for the month of February 1959 because an influenza epidemic confounds the results. The relationship between mortality and pollutant levels continued during this month, but the mortality deviations appear exaggerated compared to previous months. Significantly, the peak daily mortality deviation during the epidemic, as well as the entire winter (+ 150 deaths), occurred on the day of peak smoke (1600 $\mu g/m^3$) and SO_2 (0.6 ppm) levels for the month. On this same day sulfuric acid levels at a central London site reached 350 $\mu g/m^3$ (Commins and Waller, 1967). The peak daily deviation for the rest of the winter, even with similar pollution levels was about + 60.

Although the influenza epidemic was undoubtedly the major factor, the clinical results showing pulmonary functional changes in influenza patients with exposures to a normally innocuous neutral salt, (sodium nitrate, Utell et al., 1980) support the suggestion that the more irritant London aerosol may have been causally involved in increasing mortality during the epidemic. Such enhanced responses in influenza patients might also occur at lower concentrations, and suggest that inclusion of such epidemics in daily mortality analyses might be appropriate.

An exploration of daily mortality in London extended to 14 winters, (1958/59 to 1971/72, Mazumdar et al., 1981) provides a much larger, and thus more reliable, data base, and may be of more relevance to current conditions. This 14-year study period saw a major change in London's pollution mix, with declines in SO_2 and BS of 50% and about 80%, respectively, as well as a change in the nature of the particulate complex from the black tarry matter of the 1950's and early 1960's. After adjusting for meteorological variables, seasonal and yearly trends, and multi-collinearity of the pollutants, a strong association between daily mortality and daily pollution for the entire period was found. The authors concluded that: 1) this association was primarily due to smoke and not SO_2 and no synergistic effects between SO_2 and smoke could be shown; 2) when "episodic" days, classified as a day when BS concentrations exceeded 500 $\mu g/m^3$ plus the seven adjacent days on each side, and "non-episodic" days, which made up the balance, were analyzed separately, the association persisted throughout both classes of days, and; 3) linear and quadratic models of exposure-response between daily smoke and daily mortality were both compatible with the data.

Based on the exposure-response curves derived from these models (depicted in Figures 14-2 and 14-3, CD), the criteria document concludes: 1) steadily rising increases in excess mortality were associated with increasing BS concentrations over 500 μg/m^3, 2) under 500 μg/m^3 the relationship is uncertain, but the data provide some indications of small increases in mortality in the range of 150-500 μg/m^3 BS (CD, p. 14-21) and, 3) it is unclear whether the Mazumdar et al. analysis truly distinguished the effects of BS from SO_2 or whether the associations are due to some other unmeasured covarying factor (CD, p. 14-24). Comparison of the Mazumdar regression analyses (both linear and quadratic models) with the Ware and EPA reanalyses of the Martin and Bradley (1960) data (CD, Appendix 14-D) indicate that the four analyses were reasonably consistent in both direction and magnitude.

Other time-series analyses of daily variations in mortality and pollution in New York City suggest weak but positive associations between nonepisodic mortality and daily levels of PM (daily peaks up to 5.0-6.0 CoHs, Schimmel and Murawski, 1976; Schimmel, 1978). According to site-specific mass calibrations of particulate density readings in New York during this period, these CoH values were approximately equivalent to 575-750 μg/m^3 BS (Ingram and Golden, 1973). Although these results appear to be consistent with those from the London studies, the reliance on a single, central Manhattan monitoring station as an estimate of pollutant exposures for the entire New York City area precludes evaluation of quantitative effect levels.

b) Morbidity

The work of Lawther et al. (1970) is widely regarded as among the most reliable epidemiological investigations of the effects of particles and sulfur oxide pollution. Day-to-day changes in the health status among a group of 180 and later about 1000 chest clinic patients (mostly bronchitics, but some patients had asthma or emphysema) were shown to depend on daily variations in London pollution, measured at seven sampling sites, most close to ground level, in a ring around the city's inner residential area. The same seven sites were referred to earlier in the London mortality analyses. Although exposures varied, the pollution levels derived from the measurements in central London were considered reasonable estimates of exposure, since most subjects worked in the central district. For two winters, 1959-60 and 1964-65, the minimum daily pollution level leading to exacerbations among this group of bronchitics is considered to be about 250 $\mu g/m^3$ BS together with about 500-600 $\mu g/m^3$ SO_2 (CD, p. 14-28). As the criteria document (CD, Table 14-7) points out, however, a summary of results in a subgroup of sensitive patients for the winter of 1967-68 (Table III, Lawther *et al*., 1970) indicates a statistically significant correlation between smoke and symptoms in a winter with only one day of BS $\geq$ 250 $\mu g/m^3$ and $SO_2 > 500$ $\mu g/m^3$. Because daily temperature, SO_2, and BS levels were all significantly correlated with symptoms during the winter, the pollutant or combination of pollutants responsible for the observed effects could not be determined. Interestingly, sulfuric acid was strongly correlated with effects in a sample of patients during one winter, but less so the next.

The authors concluded that the reported aggravations in health likely reflected the effects of brief exposures to the maximum concentrations occurring during the day. These peaks were probably several times the 24-hour averages, but, because of the wide dispersion of subjects and the variation in the magnitude and timing of such peaks across the study area, the impact of peak values could not be directly examined. The smoke shade data were obtained from the same network as used in the mortality studies. A centrally sited unit not in the network was calibrated with mass readings and according to the criteria document, this calibration tends to conform "reasonably" well with the mass calibrations in the seven station network from 1959-63 (CD, p. 14-18). After that time, the calibration may have tended to overstate the true ambient mass levels (CD, Table 14-7).

Two other studies used hospital admissions/visits as indicators of acute morbidity. Martin (1964) found that applications for hospital admissions for cardiovascular and respiratory conditions among London men (45-79 yrs) were highly correlated with particulate matter (BS) and SO_2 during two winters of periodic fogs and minor pollution episodes. Again, no clear threshold exists; where deviations from the adjusted 15-day moving average become positive, however, marked increments in hospital admissions can be discerned, but only at concentrations above those apparently associated with excess mortality. Hospital admissions suggest a relatively serious effect and are clearly not as sensitive an indicator of pollution related morbidity as that used by Lawther et al. (1970). Because of these and other limitations in the use of hospital admissions data (Bennett, 1980), the Martin (1964) results do not provide reliable estimates of morbidity effects levels.

Samet et al. (1981), related hospital emergency room visits during March, April, October, and November of 1974-1977 to daily levels of TSP, SO_2, NO_2, CO and O_3 in Steubenville, Ohio, a heavily industrialized town. After adjusting for meteorological variables, weekly, seasonal and yearly cycles, possible day-of-week effects, and multi-collinearity of the pollutants in a quartile analysis, emergency room visits for all respiratory diseases did not vary significantly with pollution. However, the largest deviations from average numbers of visits occurred at the highest pollutant levels ($\geq$ 202 μg/m^3 TSP and $\geq$ 121 μg/m^3 SO_2). A regression model utilized to test for linear relationships identified a significant same-day effect of both TSP and SO_2 on respiratory disease visits. This effect was quite small, only 1% of these visits could be explained by changes in TSP and 1% by SO_2. Based on the model, the authors suggested that an increase from the observed daily mean TSP level (156 μg/m^3) of 100 μg/m^3 might result in a corresponding 3% increase in emergency room visits for respiratory disease. Nevertheless, no level at which increased visits occurred can be derived from this study.

The use of emergency room visits as a health end-point is analagous to hospital admissions data. Neither would be expected to be as sensitive an index of morbidity as that used by Lawther _et al_. (1970). The Samet _et al_. (1981) work does indicate that effects may occur in groups other than bronchitics.

2. Chronic Exposures

The relationship between long-term exposure to air pollution and health has been extensively studied, but few of the studies provide sound data or consistent findings sufficient to make quantitative conclusions. Geographic comparisons of morbidity (see Appendix B.5) and mortality rates among populations suffer from many limitations (see CD, Sections 14.1.1.1 and p. 14-35). Those studies that are useful in delineating quantitative relationships between particulate matter and chronic morbidity effects are summarized in Table 5-5.

a) Children

As pointed out in Section V-C, children who suffer respiratory insults may be predisposed to developing chronic respiratory diseases as adults. Therefore, results on children should be considered as particularly important.

TABLE 5-5. SUMMARY OF EPIDEMIOLOGICAL STUDIES PROVIDING MOST USEFUL CONCENTRATION/RESPONSE INFORMATION FOR LONG-TERM PARTICLE EXPOSURE

Area or Year	Pollution ($\mu g/m^3$) PM (BS or TSP)	SO_2	Population	Observed Effects	Levels Where Effects Are Likely (CD, Table 14-8) PM (BS or TSP)	SO_2	Comment	Study/ Location
1 2 3 4	97 BS* 230 BS 262 BS 301 BS	123 181 219 275	413;5 yrs. 194;5 yrs. 130;5 yrs. 82;5 yrs.	Higher prevalence of upper and lower respiratory tract disease in areas 2-4. Lower FEV and FVC in area 4 than in areas 1-3.	230-301 BS*	181-275	Pulmonary function tests might be questionable (methodology not described).	Lunn et al., 1967 - Sheffield, U.K.
1961 1967	180 131	Mod. Low	≈1000;25-74 yrs. ≈1400;25-74 yrs.	Prevalence of respiratory symptoms lower in 1967 than in 1961. Slight improvement in PEFR and FVC in 1967.	180	-	.1961 aerometry data based on 2 month average. Lead candles used for SO_2 measurements. Socioeconomic, smoking, other controls were applied.	Ferris and Anderson, 1962; Ferris et al., 1971; 1976 - Berlin, NH
1973	80	Mod.	≈1500;25-74 yrs.	No change in respiratory symptoms or lung function in 1973.	80-131	-		
1 2	60-150 39	20-? 20-?	1332; 7 yrs.+	No difference in lung function or chronic bronchitis but cough, phlegm, dyspnea, more common in Area 2. Asthma more common in Area 1.	N.A.		Well designed. Essentially negative study but some suggestion of pollution impact. Effects, if any, may be related to past exposures (1965-1973 mean TSP=110 $\mu g/m^3$).	Bouhuys et al., 1978 - Connecticut

*BS levels here (as $\mu g/m^3$) are crude estimates given ambiguities regarding the use of site-specific calibrations (CD, Table 14-8).

Lunn et al. (1967, 1970) studied respiratory disease and lung functions of school children in four areas of differing pollution levels in Sheffield, England. At ages 5-6 both chronic upper respiratory infections and lower respiratory tract illness were associated with residence in more polluted areas. An illness gradient corresponding to increasing pollution was found and was independent of socio-economic factors, although parental smoking, parental respiratory diseases, and home heating systems were not taken into account. Lung function ($FEV_{.75}$, FVC) was lower among the children in the most polluted area compared with the other areas. Based on the results from the lung function tests, the authors concluded that, "a pattern of respiratory disability had appeared at an early age and was sufficiently established to persist although the factors of pollution and infection were temporarily absent or at a low level." The criteria document notes that the effects levels listed in Table 5-5 have been widely accepted as being valid by several reviews (CD, p. 14-47). However, the levels must now be considered only as very approximate "observed effects" levels due to the uncertainties associated with estimating PM mass based on BS readings (CD, Table 14-8).

After four years of improving air quality, differences in the incidence of respiratory illness and in lung function among these children, now nine years old, were no longer significant (Lunn et al., 1970). The authors attributed the disappearance of differences over this period to the reduction in air pollution, especially in smoke levels with a small decrease in SO_2 levels. Given the size of the population, it is not likely that the study had sufficient power to detect small changes among the cohorts. Also, no site-specific calibrations of the BS mass readings were made at the time of the later study. Because of these limitations, the criteria document concludes that smoke levels at which no differences in respiratory disease symptoms or in lung function among the children occurred cannot be determined.

b) Adults

A series of investigations followed changes within Berlin, N.H., adults over a 12-year period (Ferris and Anderson, 1962; Ferris et al., 1973, 1976). After an essentially negative cross-sectional survey in the winter of 1961, the population was followed up in the summer of 1967 when pollution levels dropped. Small reductions in respiratory disease symptom rates, principally chest colds, together with an indication of small lung function improvement (as measured by FVC and PEFR), were associated with a decline in TSP levels from 180 to 130 $\mu g/m^3$ and in sulfur pollution levels, which were apparently moderate (based on unreliable measurements using "lead candles"). Although the TSP level for 1961 was based on only two months of monitoring, levels were clearly higher in 1961 than in 1967. Another survey in 1973 when TSP levels dropped further to 80 $\mu g/m^3$ and sulfur pollution slightly increased (based on lead candle and limited SO_2 data) found no further improvements in respiratory health. Because the trend in SO_2 did not correspond with the changes in the respiratory health of the population, the observations apparently reflected the effects of reduced particulate pollution.

Although seasonal factors were not directly controlled between the first two surveys, the investigators attempted to rule out possible confounding effects by retesting some subjects in the winter and the summer in 1961. No significant differences were found. In addition, the survey regarding respiratory illnesses was in reference to the previous three years, which likely minimized seasonal effects on illness and symptom differences between the years.

The results from this study must be interpreted with caution inasmuch as air pollution in Berlin, N.H., is dominated by the emissions of a pulp mill, unlike more typical situations in the U.S. However, the findings are generally consistent with those of other studies and suggest that marginal decrements in respiratory health and mechanics occurred between 130 and 180 $\mu g/m^3$ TSP (annual average) and that the range of 80-130 $\mu g/m^3$ TSP (annual average) is a possible lower limit at which any effects are difficult to detect.

A comparison of residents in two communities in Connecticut with different ambient exposures to TSP and SO_2 found no differences in prevalence rates of chronic bronchitis symptoms among white adults (25 years of age and older) nor in lung function among persons seven years of age and older (Bouhuys et al., 1978). A history of bronchial asthma was significantly higher among residents in the cleaner town, Lebanon. However, the prevalences of cough, phlegm, and dyspnea (when hurrying on level ground or walking uphill) were significantly higher for adult non-smokers in Ansonia. Prevalence of cough and phlegm in this group increased along a gradient from lifetime rural, to partly urban, to lifetime urban residences. The prevalence of recent wheeze was also higher in Ansonia residents, but only in comparison to lifetime rural residents in Lebanon, while part-time urban residents in Lebanon and Ansonia had equal prevalence rates as lifetime urban residents.

Response rates differed markedly between the two towns (56% versus 80%), however, follow-up analysis of the non-responders indicated that little potential for bias existed. Seasonal effects were not accounted for, but as in the Ferris study, these may have been minimized by the nature of the survey questions. The authors state that occupation was controlled for, though no data are provided to support this.

Although no differences in lung function or bronchitis prevalence were detected, the significantly increased symptom rates among the Ansonia residents indicate some small differences in respiratory health associated with pollution. It is not clear whether the reported effects related to then-current or historical pollutant levels. As the criteria document notes (p. 14-45), average annual TSP levels for the previous seven years in the urban area were between 65 and 150 $\mu g/m^3$. This is perhaps best characterized as the median value of about 110 $\mu g/m^3$. TSP levels in the rural area were provided for only one year (40 $\mu g/m^3$) but were reported to be consistently low. SO_2 was low in both areas in 1973, but might have been higher in previous years in the urban area. The lack of a detectable difference in lung function accompanied by some contrast in symptom rates is consistent with other studies (Ferris et al, 1976, 1979; Aubry et al., 1979) in which effects were either undetectable or marginal at annual TSP levels between 60 and 130 $\mu g/m^3$.

As the criteria document notes (p. 14-45), size-fractionation of a limited number of TSP samples in Ansonia by Hosein et al. (1977) showed that 81% of the TSP sample was 9.4 μm or less in diameter. This estimate, however, was based on 15 samples taken only on "dry days" with a cascade impactor of uncertain reliability. The size data were apparently characterized as log-normal, suggesting particle bounce problems and underestimation of the "MMD" common to much impactor data.

Other studies of community health provide mainly qualitative support for the existence of exposure/response relationships between sulfur oxide/ particle pollution and chronic morbidity (see Appendix B). Notable examples include two well conducted British studies (Douglas and Waller, 1966; Lambert and Reid, 1970) that indicate positive gradients of respiratory illness with increasing coal consumption. Because of various deficiencies in design or methodology, these studies cannot be considered useful in yielding information on levels of pollution at which the observed effects occurred.

VI. FACTORS TO BE CONSIDERED IN SELECTING PRIMARY STANDARDS FOR PARTICLES

This section, drawing upon the previous summary of scientific information, enumerates the key factors that should be considered by the Administrator in deciding what particle fractions should be used as an indicator of particulate pollution, establishing the levels of primary standards and designating appropriate averaging times and frequency criteria. Preliminary staff recommendations on the most appropriate policy options in each of these interrelated areas also are presented.

A. Pollutant Indicator(s)

1. Major Approaches

Faced with obvious evidence of adverse health effects in heavily polluted areas in the decades after World War II, public health authorities in the U.S. and Great Britain pushed for reductions in general particulate matter as indexed by available monitors, despite the fact that the identity of causative agents and mechanisms was not well-known. That these efforts were largely successful is evidenced by the review of epidemiological data; it is extremely difficult to reliably detect particle related health effects in areas where levels approach the current U.S. primary standards. Furthermore, as Section IV indicates, concentrations of many of the more innately toxic components of particulate matter (e.g., trace metals, BaP) have declined substantially in previously polluted areas. This has reduced the need to consider additional regulations (ambient or source) for control of such substances. The initial steps to control particles largely undifferentiated in terms of size and chemical composition appear to have been good public health policy, even though "particulate matter" is toxicologically undefined.

Nevertheless, the efficiency and effectiveness of total particulate standards have been justifiably questioned in recent years. In this standards review, then, it is necessary to use available information to specify what measurable particulate fractions are the most suitable indicators of those particles potentially of concern to health; in essence, we must define the pollutant.

Although a number of approaches are available, the major policy options with respect to particle indicators include:

1) Indicators used in community epidemiological studies (TSP, BS, CoH);
2) Chemically specific indicators - (By class, e.g., sulfates, or compound, e.g., sulfuric acid);
3) Size specific indicators (e.g., FP, MRP, BMRC, IP); and
4) Combined particle (as above)/SO_2 index.

Each of these approaches is discussed and evaluated briefly below.

a) Indicators used in Community Epidemiological Studies

Three major indices of particulate matter have been associated with health effects in community epidemiological studies: Total Suspended Particulate Matter (TSP), British Smoke (BS), and Coefficient of Haze (CoH). The advantages of using these techniques as indicators include direct relationship to the quantitative health data base and/or historical continuity of particle data. None of these approaches, however, were designed to account for respiratory tract deposition or variable ambient composition. Each has specific design flaws making application for U.S. health standards much less than optimal. Briefly, major problems include:

i) TSP - Windspeed dependent particle acceptance of the hi-vol sampler (< 25-45 μm) is unrelated to respiratory tract deposition. In current U.S. atmospheres, substantial quantities of coarse particles too large to reach the thoracic region are collected. Thus, large (> 10 μm) coarse particle control is emphasized.

ii) BS, CoH - Particle acceptance of these instruments (< 4-5 μm) includes much of the material potentially deposited in the alveolar region of the lung, but may exclude some alveolar deposition, and a major fraction of particles that can be deposited in the tracheobronchial region. More importantly, the BS reading varies more with darkness of the particles (carbon content) than with mass, making associations with mass highly site and time specific. Control of primary elemental carbon emissions is emphasized; elemental carbon is a minor contributor to fine and total mass in current U.S. atmospheres.

iii) TSP/BS Relationships - In addition to the above problems, the lack of any consistent relationship between TSP mass and BS reflectance diminishes one of the major advantages of these indicators; direct relatability to the available quantitative health data.

b) Chemically Specific Indicators

Regulation of specific chemical substances or identified classes could potentially improve the effectiveness of particulate control programs. Ideally, it would be useful to identify a few specific chemical substances of defined size range that are responsible for the vast majority of observed effects. Despite efforts on three continents over the last thirty years, however, no such specific "active agents" have been unequivocally identified. Instead, our review suggests that a large number of different particulate substances may produce responses in animals and humans, usually at levels higher than those found to be associated with effects in community epidemiological studies.

Identification and control of each of the many ambient aerosol components would be difficult to accomplish, time-consuming, and would place excessive monitoring, compliance, and other requirements on affected agencies and industries with no clear potential for improving public health protection over general particulate regulation. Exceptions, such as trace constituents of high toxicity which are inadequately controlled by general particle standards, can be regulated by specific ambient or emissions standards, as in the case of lead or asbestos.

A somewhat more practical intermediate approach would be to use chemical classes (e.g., "sulfates", "organics") as indicators of particulate pollution. The available epidemiological data base, however, does not provide quantitative or qualitative support for chemical class standards and both animal and human laboratory studies indicate variable toxicity even within such categories. Given current knowledge and technical capabilities, separate standards for chemical classes would be difficult to support and implement, again with no obvious advantages for improving health protection. Work to identify particularly significant classes/sources/substances is needed to support future standard reviews.

c) Size Specific Indicators

All commonly used particulate monitoring devices are in some measure size specific; the key issue is to determine what the most appropriate size fractions are for ambient air quality standards. Aside from continuity with historical epidemiological instruments (see a) above), the two major approaches to selecting such size fractions include

1) size divisions according to regional human respiratory tract deposition and 2) size divisions according to typical atmospheric size distributions, for example, fine and coarse. These two approaches are not necessarily completely independent.

i) Human Respiratory Tract Deposition

Particulate indicators based on regional human respiratory tract deposition have long been adopted in occupational settings (Lippmann, 1970) and were recently recommended for ambient particles (Miller _et al_., 1979; ISO, 1981). Although this approach forms the most logical basis for defining a health related particulate standard, there may be difficulties in relating deposition derived indicators to the epidemiological results, as well as uncertainties in translating available human deposition data into uniform size fractions. Criteria are discussed more fully in Section 2 below.

ii) Ambient Size Distribution

Recognition that ambient particle mass and volume are typically distributed bimodally has led to the suggestion that the health (as well as other) effects of the two modes might ultimately be treated separately (NAS, 1977a; Miller _et al_., 1979). Because the chemistry of the two modes tends to differ, this approach is essentially a simplified version of the chemical class index approach discussed above.

This "natural" dividing line is at least partially relatable to human deposition. The typical minimum dividing the fine and coarse modes (1 - 3 μm) is near the 50% size "cut" used in occupational settings to indicate insoluble "respirable" particles (3.5 - 5 μm).* Significant alveolar penetration, the enhanced effects with decreased particle size noted in some animal studies, and the substantial surface area in the sub-micrometer range have long been cited as indications of the need to develop data in support of separate fine particle standards (Natusch and Wallace, 1974; NAS, 1977a). Moreover, it has been argued that the British epidemiological studies might be relatable to a fine particle index, both because of the nature of the BS measurement and the nature of the historical British aerosol (Waller, 1980).

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*Although precise collection methods may vary, "respirable" particles have been defined as those penetrating to the non-ciliated portions of the lung (Miller _et al_., 1979).

Nevertheless, some difficulties and uncertainties exist in adopting a fine/coarse fractionation for primary standards. Briefly, these include:

1) The dividing line between fine and coarse particles is neither sharp nor fixed and substantial over-lap may occur.
2) The minimum in the bimodal distribution may, in effect, disappear near strong sources or under other peak loading conditions (see Section IV); this minimum as well as smaller coarse particles lie near the size range of maximum efficiency for alveolar deposition (2 - 4 μm).
3) Although the two modes have differing origins and chemistries, each is chemically heterogeneous.
4) The respiratory tract in effect alters the ambient distribution, with a mixture of fine and coarse particles being deposited in the tracheobronchial and alveolar regions.
5) The mixing of modes in the respiratory tract and the heterogeneity within each mode blur the distinction between the modes in terms of health effects. For example, with respect to health effects, fine sulfates may have more in common with coarse nitrates than with fine polycyclic organic compounds. Similarly, it is not clear that 1 μm carbon particles are more likely to result in pneumoconiosis than 4 μm silica particles.

6) Respirable sampling used in occupational studies is apparently not a good indicator of some effects associated with occupational exposure to insoluble mineral dusts, particularly industrial bronchitis (Morgan, 1978). The fine fraction alone, then, also would not adequately include particles related to such effects.

7) Given that coarse "insoluble" dusts can result in responses such as bronchoconstriction, altered clearance and alveolar tissue damage (Section V), and the likelihood that coarse particles were present in British air (Section IV), it would be premature to ascribe all of the effects noted in British studies solely to fine particles. Neither the British nor other community epidemiological studies permit unequivocal separation of the effects of fine and coarse particles. Indeed, until ongoing studies make more definitive divisions possible, the most prudent and defensible course may well be to combine both British and U.S. epidemiological data in support of a single size specific indicator based on human deposition data.

d) Combined PM/SO_2 Index

Elevated particle levels associated with health effects in historical episodes were most often accompanied by high levels of sulfur dioxide. As discussed in previous sections, it has been difficult to separate the effects of particles and SO_2 in these cases, and both pollutants might have been acting as indicators for some hitherto unidentified "active agent." Mechanistically, particles may increase penetration of sorbed SO_2 or otherwise enhance toxicity by chemical or physical transformations. Combined SO_2/PM indices have been suggested to account for these potential interactions.

The relevance of such a combined index to contemporary U.S. atmospheres is, however, unclear. The sources of high SO_2 levels often are well-controlled with respect to particulate matter while many major sources of particles emit little or no SO_2. The combination of high levels of both SO_2 and particles is comparatively rare, since the SO_2 standards are largely met. Moreover, particles may interact with other gases more commonly found in U.S. atmospheres than in historical epidemiological studies, notably photochemically generated pollutants such as ozone, and NO_2.

Epidemiological evidence suggests that reduction in particulate matter levels may improve health status even when SO_2 levels remain elevated or are initially low (Lawther _et al_., 1970; Ferris _et al_., 1973). As Lawther _et al_. (1970) point out, this should _not_ be construed as an indication that SO_2 produces no effect, merely that control of one indicator can reduce risks.

In light of the above considerations, linking the allowable U.S. particle levels to SO_2 concentrations through a combined index would not necessarily offer any improvement in health protection over a separate particle standard. Even with a separate particle standard, the possibility of linking SO_2 standards to particle levels still can be considered later in the review of the SO_2 standard.

2. Staff Recommendations for a Particle Indicator

a) Recommendations for Approach

Based on the above evaluation, the EPA staff reaches the following conclusions with respect to an approach to selecting a particle indicator:

1) The current primary standard indicator, TSP, can and should be improved upon during this standard review. Choice of other indicators used in historical epidemiological studies (BS, CoH) is not advisable, both on health and technological grounds.

2) Movement to chemically specific indicators ultimately may be desirable but not advisable during this review. Of the particulate chemical classes, most is known about sulfates. Nevertheless, most reviews have concluded that insufficient data exist to set a separate national sulfate or sulfuric acid standard (NAS, 1977a, 1978). We concur with these judgments.

3) From the standpoint of protecting public health from the effects of particulate matter, a combined SO_2/PM index is not necessary.

4) Choosing a size specific particle indicator based primarily on respiratory tract deposition data is the best approach for improving the particulate matter standard. The multimodal nature of the ambient aerosol distribution should be considered, but should not form the principal basis for selecting a health-related indicator at this time.

b) Recommendations for a Size Specific Indicator

The main questions to be addressed in selecting a size specific indicator are i) what particles deposit in various regions of the respiratory tract; and ii) what are the potential health consequences of such deposition for normal and sensitive population groups? Drawing on the discussion in Section V of available regional deposition data as applied to various ambient size distributions, potential mechanisms of toxicity and possible health responses, a brief summary of key considerations for each region is outlined below.

i) Extrathoracic Region - The only particles uniquely deposited in this region are coarse mode particles larger than 10 - 15 μm. Because clearance is normally rapid (minutes), only systemic toxicants (not normally present at high levels in this size range), direct irritants, or pollens are likely to produce effects. The major responses are symptoms such as rhinitis and dryness in the throat. Occupational exposures suggest wood dusts can slow clearance and may produce cancer, but no evidence suggests that inert dusts or other common ambient coarse particles produce these effects.

ii) Tracheobronchial Region - Particles deposited in this region (<15 μm) are also deposited in one or both of the other regions. Average clearance takes hours to days, but may be slower at the airway bifurcations. Potential physiological responses to high levels of both coarse "inert" dusts and acid aerosols include bronchoconstriction, altered clearance, and buildup of fine and coarse particles at the bifurcations. These responses may result in reduced lung function, aggravation of existing respiratory disease (particularly in bronchitics and asthmatics with enhanced tracheobronchial deposition), increased infectious disease and potentiation of the effects of cigarette smoking or other exposures in development of bronchitis and the most common forms of lung cancer. Although many of these responses have been observed in community epidemiological studies, the relative role of fine and coarse particles in producing them is not clear.

iii) Alveolar Region - Particles less than 10 μm may be deposited in this region. Because clearance of insoluble materials may require months to years, any material deposited is of concern. Possible responses include reduced lung function, damage to lung tissues, increased susceptibility to infection, and aggravation or potentiation of cardiopulmonary diseases. Although more of the total deposited mass in this region is likely fine mode, much of this is soluble and cleared rapidly by dissolution. In current U.S. atmospheres, over half of the insoluble material deposited in this region likely originates in the coarse mode. Autopsies of groups exposed to fugitive dusts suggest potential for damage from such deposition.

Based on the above considerations, it appears that the risk of adverse health effects associated with extrathoracic deposition of commonly found particles larger than 10 to 15 μm is sufficiently low that they can be safely excluded from the primary particulate standard. In rare cases where systemic toxicants are present in significant concentrations in this fraction, any general particle standard likely would be insufficient and other sections of the Clean Air Act (111d, 112) would have to be considered. Pollens, although included in TSP samples, have not generally been subject to particulate pollution control programs, so their exclusion would not materially affect health protection.

Our review suggests that the risks of adverse effects associated with deposition of coarse particles in the tracheobronchial region are markedly greater than those for extrathoracic deposition. Support for this position can be found in the International Standards Organization

(ISO) report (1981) which states "It was the opinion of the ad hoc working group that tracheobronchial deposition is more often of health interest than extrathoracic deposition." Until the chemical and physical characteristics and biological effects of coarse particles and accompanying trace constituents are better understood, it would be premature to exclude those deposited in this region from control. It follows that fine particles, particularly hygroscopic particles, both acid and neutral, depositing in this region also should be included in a particle indicator.

The risks of fine and coarse particles deposited in the alveolar region also are of clear concern. Nevertheless, because substantial overlap exists between particle deposition in the tracheobronchial and alveolar regions and because epidemiological studies frequently do not permit distinction between these regions or between the effects of fine and coarse particles deposited in these regions, separate indices for alveolar and tracheobronchially-deposited particles are not advisable at this time. Rather, an index representing particles that penetrate to the thoracic region or "thoracic particles" (TP)* is recommended. This indicator includes both alveolar and tracheobronchial penetration.

The approach and criteria for defining TP advocated by Miller et al. (1979) and utilized by the International Standards Organization (ISO) are consistent with Clean Air Act guidance for protecting sensitive populations and providing a margin of safety. Specifically, size specific deposition estimates should reflect those portions of the population who through illness, habit, exercise or other conditions breathe through the mouth.

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*The term "thoracic particles" ('TP') will be used throughout the remainder of this paper to mean particles penetrating to the thoracic region. The term is intended for use only in the context of this staff paper to distinguish it from other particle indicators associated with specific size cuts, namely inhalable particles (<15 μm), respirable particles (3.5-5 μm), and fine particles (<2.5 μm).

Using available data for mouth only deposition, the ISO has essentially recommended two alternative definitions for particles potentially depositing in the thoracic region; those particles less than 15 μm and 10 μm respectively. In effect, TP monitors would need a 50% "cut" point at one of these diameters with a specified geometric standard deviation (GSD) or other indicator of sharpness. Both are based on extrapolation of extrathoracic deposition data for mouth breathing which suggests that under normal flow conditions, less than 10% of 15 μm or larger particles reach the tracheobronchial region. The 15 μm definition of TP represents the precautionary "envelope" approach advocated by Miller _et_ _al_. (1979) in defining "Inhalable Particles". The 10 μm definition of TP represents the more traditional approach of approximating the intake deposition profile of the respiratory tract, in this case, the thoracic region. Although particle deposition patterns vary with factors such as individual characteristics and flow patterns, based on controlled experiments involving mouthpiece inhalation (Figure 5-1), roughly 50% of inspired particles of 10 μm size are deposited in the tracheobronchial region with mouth breathing.

The staff believes that both definitions have merit. Some considerations in this regard include:

1) These alternatives happen to lie near the peak of the coarse mode (see Figure 4-3). The major difference will be in the amount of coarse material collected.
2) The 15 μm limit ensures almost all tracheobronchial deposition and additional extrathoracic deposition (< 5 μm) are included, and, in that sense, is more conservative. The 10 μm limit may be somewhat conservative for thoracic deposition because the

data are based on mouthpiece experiments. The use of mouth pieces should tend to increase particle penetration. The "margin of safety" however, is a function of both the size fraction and level of the standard. Thus, a 15 μm cut does not inherently provide any greater "margin of safety", than a 10 μm cut.

3) The probability of thoracic deposition, time for clearance, and risk of health effects increases as particle size is decreased from 15 μm to 10 μm.

4) Both definitions offer an envelope for particles penetrating the alveolar region. This is a desirable feature, in that the full range of alveolar penetration (up to 10 μm) is included in the sample.

5) Based on indoor/outdoor studies (CD, pp. 5-127-131), the 10 μm fraction may be a more reliable indicator of that portion of ambient particles that contributes most to indoor exposures.

6) Either definition would tend to place greater weight on controlling smaller particles than does a TSP standard.

7) The availability of 10 μm monitors and ambient data for regulatory analyses is not a serious constraint on this decision. Prototype samples and inlets for retrofitting the existing IP (15 μm) network are available and data from the current size specific networks can be interpolated to provide reasonably reliable estimates of mass <10 μm for interim use. Theoretical and practical experience to date would suggest that 10 μm samplers would be more reliable and less sensitive to windspeed than 15 μm samplers (CD, p. 2-61).

8) The representatives of member countries of the ISO recently voted to adopt the 10 μm definition for measuring the thoracic fraction of particulate matter (Ogden, 1981).

Given these considerations, the staff recommends that the 10 μm alternative be selected for the particle standard indicator. This alternative appears to be at least somewhat conservative with respect to thoracic penetration and, all else being equal, offers the possibility of more reliable monitoring. Moreover, matching the intake profile of the thoracic region places greatest emphasis on fine particles, while including those coarse particles most likely to be of health concern. Based on typical urban particle size distributions, fine particles constitute over 60% of the particle mass less than 10 μm. A 10 μm particle sampler would collect 100% of the fine mass (< 2.5 μm) and between 50 and 100% of the coarse mass in the 2.5 to 10 μm range. In contrast, the deposition data of Figure 5-1 show that most fine mass is not deposited in the respiratory tract, while for mouth breathing most coarse mass in the 2.5 to 10 μm range is deposited. Given the larger surface area in the fine mode as well as other concerns outlined previously, the greater weight given fine vs. coarse particles by a 10 μm indicator appears to be prudent and appropriate.

In addition to selecting a particle size, practical tolerance limits for both the midpoint and shape of a sampler particle acceptance curve must be specified. At 10 μm, it is possible to design sharp (low GSD ≈1.1-1.3) or somewhat less steep particle inlets. The sharper acceptance curves tend to more accurately reflect the ambient concentration, while less steep curves appear to more closely conform to the ideal of matching respiratory tract characteristics. An EPA analysis of sampling alternatives has been conducted in support of this staff paper

(Rodes et al., 1981). The following conclusions and recommendations are derived from that analysis:

1) The respiratory tract penetration/particle size data are not lognormal and hence a single GSD is not realistic. The Chan and Lippmann (1980) data provide a reasonable basis for an "ideal" sampler that matches respiratory tract penetration.

2) Recommended sizing tolerance limits for TP samplers are $D_{50} = 10 \pm 1$ μm for all windspeeds from 2 to 24 km/hr. No exact constraints are given for sharpness of cut. However, the total mass collected by the sampler should be in agreement with that collected by the "ideal" sampler for expected ranges of windspeed and particle distributions.

3) At least two prototype sampling systems are available that are expected to produce mass concentration results within ± 10% of the ideal TP sampler and within 15% of each other under "worst case" fugitive dust sampling conditions.

In summary, the staff recommends that the particle standard indicator represents those particles penetrating the thoracic region. The size range should include those particles less than a nominal 10 μm and sampler performance criteria should be related to respiratory tract deposition data.

B. Averaging Time and Form of the Standard

1. Averaging time(s)

The current averaging times for the particulate matter NAAQS are annual and 24 hour and were based on available epidemiological studies. The quantitative and qualitative studies outlined previously still suggest the need for both a 24 hour and annual standard. Some epidemiological investigators (Lawther et al., 1970) have speculated that the observed health effects might be largely due to short-term peaks on the order of an hour. Controlled human exposures to specific particles also indicate that some effects can occur after exposures of minutes (e.g., Utell et al., 1980) to hours (McDermott, 1962). Controlled human data on specific substances, however, do not provide a satisfactory quantitative basis for selecting general particulate standards and epidemiological studies have as yet been unable to provide exposure/response information for less than 24-hour periods.

Therefore, the staff recommends retention of 24 hour and annual averaging times for particulate matter standards.

2. Form of the Standard

The current particulate matter annual primary National Ambient Air Quality Standard (NAAQS) is based on the geometric mean of all valid daily total suspended particulate (TSP) measurements in a calendar year. The use of a geometric mean in the standard may have originated from the fact that TSP and other pollutant levels do not follow a normal distribution. It may also, in part, be a carry over of its use to describe long-term air quality in the Winkelstein (1967) study, which was a major epidemiological study featured in the 1969 criteria document for particulate matter (DHEW, 1969).

The use of an annual average based on the arithmetic mean of the daily averages may provide a more appropriate form of the standard for providing health protection. Health effects are most likely a function of dosage, a quantity more directly related to the arithmetic mean (Mage, 1980). The arithmetic mean is more sensitive to repeated short-term peaks than is the geometric mean. If these peaks fall below the level of the short-term (24 hour) standard (which is designed to protect against acute effects from maximal 24-hour exposures), but above some lower level at which repeated short-term exposures can adversely affect health over the long run, the arithmetic mean can provide better health safeguards. An arithmetic mean form of the standard will also allow comparison and consistency with other annual NAAQS's. For these reasons, the staff recommends an arithmetic mean form of the annual particulate matter NAAQS.

The staff recommends that the 24 hour standard be stated in a statistical form rather than the current deterministic form (the current standard is not to be exceeded more than once per year). This would mean either 1) that the allowable number of exceedances would be expressed as an average or expected number per year, or 2) that a given percent of the daily values would be expected to be less than or equal to the standard level. The emissions reductions to be achieved in the required control implementation program would be based on statistical analysis of monitoring data over a multi-year period (e.g., the preceeding 3-year period).

The statistical form can offer a more stable target for control programs and, with reasonably complete data, is less sensitive to truly unusual meteorological conditions than the deterministic form. The general limitations of the deterministic form are discussed more fully elswhere (Biller and Feagans, 1981). Recognition of the limitations has led EPA to promulgate or propose statistical forms for the ozone and carbon monoxide standards.

An additional consideration arises in the case of particulate matter. Unlike ozone or CO, particulate matter is not monitored continuously but, typically, on a one in six day schedule. In a year where there are actually two exceedances of a standard level, the probability of detecting both is only 0.03. Thus, the relative protection provided varies significantly with frequency of sampling. This represents a flaw in both the form of and monitoring requirements for the current standard. The interaction between form of the standard and alternative monitoring requirements should be considered in developing the standard. The issue of allowable number of exceedances is an important one in that it interacts with selection of the level of the standard. This interaction should be specified so that it can be explicitly considered.

Conceptually, the least complex approach might be first to select an acceptable standard level based on one exceedance and complete sampling, and then adjust the level so as to provide roughly equivalent protection under alternative exceedance/sampling schedules. Table 6-1 illustrates the relationship between standard level and exceedances for one representative temporal distribution function (Frank, 1981) and a sampling frequency of 365/yr.

TABLE 6-1. STANDARD LEVEL AS A FUNCTION OF EXCEEDANCES

Number of Allowable Exceedances*					
1	2	3	4	5	6
1.00	0.92	0.87	0.84	0.81	0.79

*Entries are the relative levels of an expected exceedance (statistical form of the standard, 3 year attainment test) that provide the same degree of protection as provided by 365 sampling days per year with one allowable exceedance (normalized to 1.0). Values are for an assumed exponential distribution ($\lambda\theta = 2.0$) but are also close to those for lognormal (GSD = 1.6) and Weibull (K = 1.5) distributions (Frank, 1981).

The table shows that as the number of allowable exceedances increase, the level of the standard to provide equivalent protection decreases. The analysis of Frank (1981) shows that the degree of protection is not very sensitive to sampling frequencies between 60 and 365 per year for the statistical form of the standard. More complete sampling does, however, provide a more stable target. For the statistical form with one expected exceedance, as the sampling frequency is decreased from 365 to 61/yr, the equivalent standard level decreases from 1.0 to 0.98. In contrast, with the current deterministic form and the same distribution function, as the sampling frequency is decreased from 365 to 61/yr, the equivalent standard level decreases from 1.0 to 0.76 (Frank, 1981).

For the purposes of this paper, ranges for 24-hour standards presented later will assume a statistical form, with no more than one expected exceedance per year over any consecutive three year period. Alternative numbers of exceedances (and sampling frequencies) may be desirable and should be considered. In so doing, the interaction between these alternatives and the level of the standards can be specified.

C. Level of the Standard(s)

1. General Considerations

Selecting a particulate matter air quality standard with an adequate margin of safety is a difficult challenge for the decision maker. In addition to the normal uncertainties involved in making judgments on the health risks of specific substances (such as carbon monoxide or NO_2) in real world atmospheres, the decision-maker must take into account the fact that the chemical and physical characteristics of particles vary with time and location. This makes translation of epidemiological evidence difficult and diminishes the utility of controlled human and animal studies in making quantitative judgments. Some important general considerations in approaching this problem are outlined below.

1) Particulate matter remains a pollutant class with variable composition. The relative proportion of fine and coarse particles and chemical components such as sulfuric acid, carbon, and silica vary significantly with geographical location, source mix, meteorology and time. The recommended size specific indicator (TP) only partially reduces this variability, but does limit control to those particles with the greatest potential for damage. Nevertheless, the risks (margins of safety) associated with attainment of a given national standard will vary from one location to another.

2) It follows that, although the scientific literature supports the notion that various mixes of particles pose risks to health, the basis for any general particulate standard is largely a public health policy judgment. The more rigorous approach of identifying each key toxicant and combination must continue, but control of size-specific particles ultimately may continue as an effective policy response to reduce the need for more numerous specific standards.

3) As noted earlier, epidemiology, with mechanistic support from toxicological and deposition studies, provides the major basis for identifying and/or setting the level of the standard. None of the published studies have used TP as a pollutant indicator.

4) The difficulties in conducting and interpreting epidemiological studies are discussed in Section V-D and in the criteria document. All epidemiological studies appeared flawed to some degree (e.g., confounding variables, characterization of exposures), further limiting the reliance that can be placed on the results of any single study. Furthermore, discord is apparent among epidemiologists over the acceptability and proper use of some studies (e.g., Holland _et_ _al_., 1979; Shy, 1980; Ware _et_ _al_., 1981).

5) As pollutant levels decrease, it becomes extremely difficult to discern effects in epidemiological studies, indicating that pollutant effects, if any, are small in comparison to other causes. In some cases (e.g., mortality), however, even small differences in effects may be viewed as serious. In this context, it should be noted that a prospective epidemiological study would need a study group of about 200,000 people to detect a 2% (2/100) difference in mortality rate (Wilson _et_. _al_., 1981).

6) Even the best epidemiological studies can do no more than provide site, time, and monitor specific associations between levels of a given particulate/other air pollutant mix and observed health responses. Moreover, such studies often provide no clear evidence of population "thresholds." Thus, the approach of identifying specific "lowest demonstrated effects levels" and adding margins of safety considerations is less appropriate in this case. Instead, attempts must be made to assess the nature of health risks along a continuum of exposures using the full range of available health and exposure data.

7) Based on the above considerations, it is apparent that decison makers will have to deal with unusually large uncertainties in selecting national particle standards. In evaluating health risks (margins of safety), the following aspects must be balanced:

a) Particle size range selected as an indicator;
b) Level (concentration);
c) Range of expected aerosol composition; and
d) Potential effects not examined in the more quantitative epidemiological studies.

The conceptual relationship between size index and levels of the standard in the range of interest is illustrated in Figure 6-1. The range of aerosol composition initially must be considered in comparing current U.S. compositional data to the composition of particles in the relevant epidemiological studies (if available). In most cases, it may be reasonable or necessary to assume comparable toxicity due to a lack of detailed data. In addition, the quantitative community epidemiological data base does not provide exposure-response information for some of the likely effects of major TP components (e.g., pneumoconiosis from insoluble particles). Thus, the levels of interest for TP derived from the quantitative studies also should be evaluated with respect to any unquantified potential effects that may be reasonably anticipated, based on TP composition and the qualitative effects studies summarized in Section V-B.

2. Qualitative Assessment of Risks of Thoracic Particles

The most useful epidemiological studies for establishing exposure-response relationships are presented in the main text of Chapter 14 of the criteria document and summarized in Section V-D and Tables 5-4 and 5-5

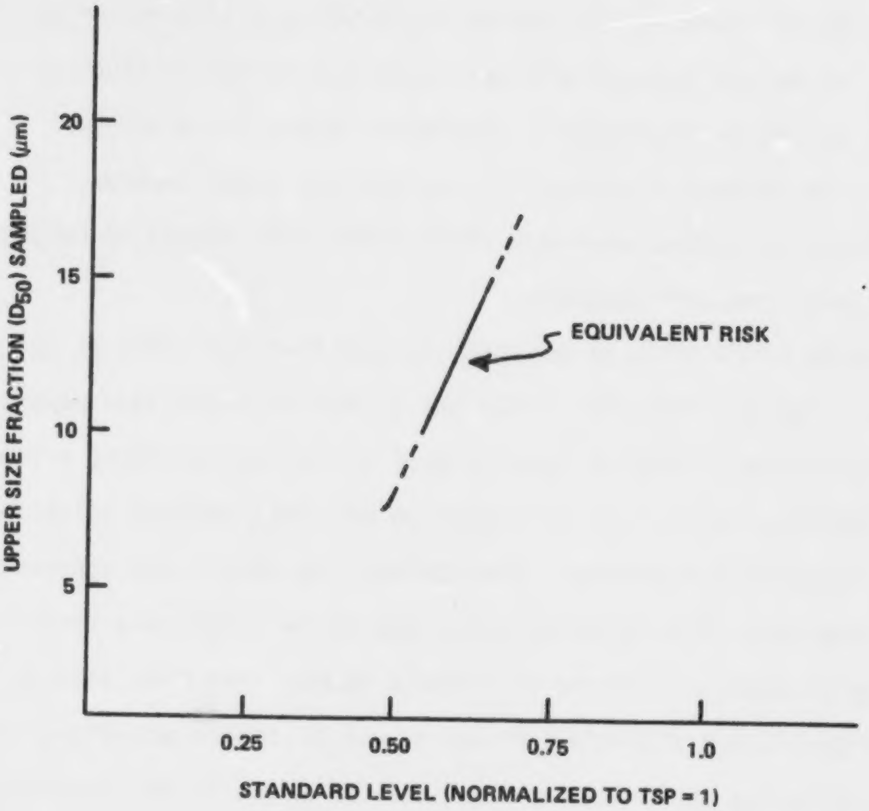

Figure 6-1. Conceptual relationship between level of standard and size indicator. Assuming a typical ambient distribution and equal toxicity in the 10-15 µm range, equivalent health protection may be achieved for <10 or <15 µm size fractions by adjusting the level of the standard as indicated. In essence, as the size of the indicator increases, a larger portion of the coarse mode is collected, permitting the standard level to increase with no change in health risk.

of this paper. Following the approach used in the criteria document and by other reviewers, the PM and SO_2 concentrations listed represent indicator levels at or above which the pollutant mix present in the particular study location is likely to be associated with observed health effects. As such, they are characterized as "effects likely levels" in Table 5-4 and 5-5. They must not be viewed as "thresholds" below which no effects can be inferred. Given the variable pollutant mixes and indicators studied, they also can not be unequivocally considered "demonstrated effects levels" in the context of current U.S. particulate matter exposures. Study authors and various reviewers often differ with respect to specifying effects levels from these studies.

As noted in the criteria document, by some reviewers (Ware et al., 1981; Wilson et al., 1981; NAS, 1978) and in Section V-D of this paper, a striking feature of many of these studies is the lack of strong evidence for any threshold levels and the suggestion of a dose response relationship along a continuum of exposures. Nevertheless, the most common approaches to assessing risks of particulate matter and sulfur oxides have been: 1) tabular or graphical listing of "effects levels" from these studies, sometimes normalized to single indicators; and 2) regression analyses of national mortality statistics, making little or no use of the remaining epidemiological data (e.g., Lave and Seskin, 1977). The first approach gives no clear sense of the risk of effects below the stated levels, even though the original data often suggest this possibility. The second approach, while usually consistent with a continuum of exposure/response even at low levels, looks only at one endpoint and often produces conflicting results with respect to pollutant indicator and strength of the relationship because of confounding variables. Although some reviewers feel that these studies suggest particle-sulfur/mortality associations at ambient levels (Thibodeau _et_ _al_., 1980; Wilson _et_ _al_., 1980), it does not appear that these studies provide reliable quantitative estimates of risk (Ware _et_ _al_., 1981). Furthermore, neither of the above approaches makes much use of the weight of evidence from relevant qualitative epidemiology, controlled human exposures, and animal toxicology.

The following sections present a brief staff assessment of the concentration/response relationships suggested by the most significant epidemiological studies in the criteria document and how these studies may be applied in developing ranges for decison-making on standards for particulate matter, as indicated by Thoracic Particles (<10 μm). The presentation also outlines a qualitative assessment of the key factors that affect the margins of safety (risks) associated with the concentration/response relationship derived from these studies, as translated to contemporary U.S. exposures. This includes identification of those important aspects of the qualitative human and animal health studies as summarized in Section V (and Appendix B) that should be incorporated into margin of safety considerations. Short and long-term exposures are discussed separately.

a) _Short-term Exposures_

i) _Derivation of Ranges of Interest from Epidemiological Studies_

1. Concentration-Response Relationships

Although a number of epidemiological investigations provide qualitative evidence for the effects of short-term exposure to particulate matter (usually with SO_2 and other pollutants), the criteria document indicates that those most useful for developing quantitative conclusions include a series of studies and reanalyses of daily mortality in London (Martin and Bradley, 1960; Martin, 1964, Ware et al. 1981; Mazumdar et al., 1981; CD, p. 14-16 to 14-24) and studies of bronchitis patients in London (Lawther et al., 1970).

The London mortality studies have been characterized by the criteria document as suggesting notable increases in excess mortality occurred in the range of 500-1000 $\mu g/m^3$ BS and SO_2 and are most likely when both pollutants exceeded about 750 $\mu g/m^3$. These estimates represent judgments of the most scientifically reliable "effects levels", for daily smoke and mortality, at least in the context of historical London pollution exposures. Because of the severity of the health end points and the need to consider margins of safety in standard setting, it is important to determine whether the data support the possibility of health risk below these "effects likely levels." As discussed in Section 14.3.1.2 of the criteria document, the London mortality studies and reanalyses do support the possibility of a monotonic dose-response relationship for particles (and perhaps SO_2). Figure 6-2 (CD, Figure 14-1) illustrates the relationship between deviations from average mortality(15 day moving mean) and smoke and SO_2 concentration for the London winter of 1958-59 (Ware et al., 1981). Figure 14D-3 in the criteria document presents unadjusted mortality data from the same winter but includes confidence intervals that show the uncertainties added by monitoring and other errors. Both seasonally adjusted and crude mortality data suggest the possibility of a gradient in mortality across a range of exposures from about 150 $\mu g/m^3$ (the lowest average daily values) to over 1200 $\mu g/m^3$ of both BS and SO_2. Analyses by Martin and Bradley (1960) indicate that, during this winter, temperature was not

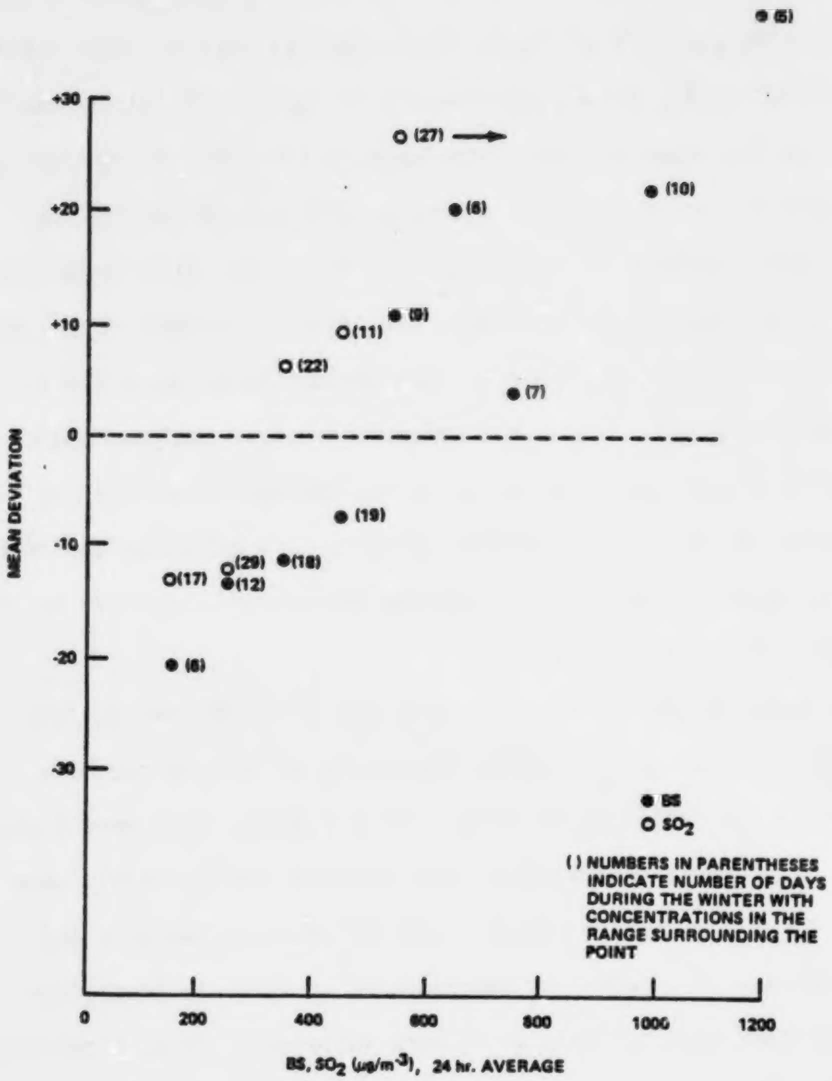

Figure 6-2. Average deviations of daily mortality from 15 day moving average by concentrations of smoke (BS) and SO$_2$ (London, November 1, 1958 to January 31, 1959). Martin and Bradley (1960) data as summarized by Ware et al. (1981). As daily pollutant levels decrease, the positive mortality deviations also decline. At still lower pollution levels, daily mortality deviations fall increasingly below the 15 day moving mean, suggesting the possibility of a continuum of exposure-response over the range examined. Uncertainties in mortality data and aerometry are not depicted by these averaged points. Examination of Figures 14D1 and 14D3 (CD) give some sense of the variability in unadjusted mortality and in aerometry. Note that point with arrow represents mortality deviation for all days with SO$_2$ >500 µg/m^3.

significantly correlated with mortality, and humidity was only marginally significant. The analysis of Figure 14D-3 suggests that at lower concentration ranges (150-500 $\mu g/m^3$), errors associated with variability in air quality estimates from the seven station London network are small to negligible. Other possible errors in aerometry are more difficult to specify (CD, p. 14-18 to 19). Because of the variability in the mortality data and the nature of the confidence intervals, the criteria document notes that the relationship becomes increasingly less certain below about 500 $\mu g/m^3$. Although the Ware et al. (1981) and criteria document analyses both omit data from a month with an influenza epidemic, the Martin and Bradley (1960) data show that the association between mortality and pollution continued during the epidemic. Peak mortality during the epidemic occurred on the day with peak smoke and SO_2.

As indicated in the criteria document (CD, p. 14-21) and by data summarized by Mazumdar et al. (1981), the winter of 1958-59 may have been unusual in the following respects: (1) mortality rates were higher than for 12 of 13 subsequent winters; (2) particle and SO_2 levels were higher than in all subsequent winters; and (3) relative humidity and the number of days of intense fog contributing to episodic conditions (with concomittant high sulfuric acid) were increased. Thus, examination of mortality-pollution relationships in subsequent years is important in determining whether any indication of health risks below the suggested effects levels persists in later years.

Analysis of the adjusted daily mortality for all London winters from 1959-72 (Mazumdar et al. 1981) again illustrates the possibility of a continuum of response over a wide range of particle concentrations (Figure 6-3a,b) (CD, Figures 14-2 and 3). Of particular interest is the fact

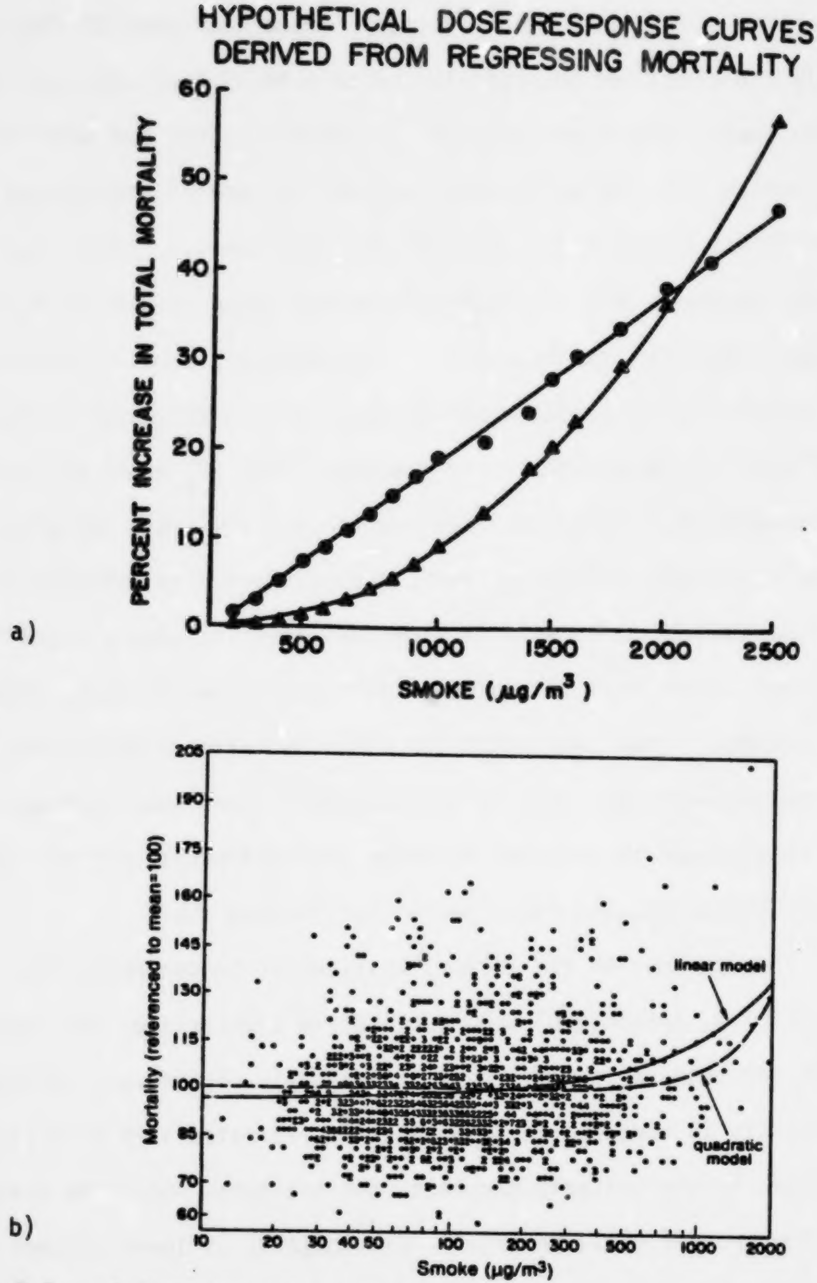

Figure 6-3. a) Hypothetical concentration-response curves derived from regressing mortality on smoke in London during winters 1958/59 to 1971/72. Results obtained with linear (●) and quadratic (▲) models are depicted for comparison. b) Same concentration and response models plotted on the scattergram of adjusted mortality and smoke for same winters as in (a). Note that the smoke scale (abscissa) is logarithmic. This plot illustrates variability in the data (Mazumdar et al., 1981).

that the investigators analyzed episodic (days of >500 μg/m^3 and adjacent 7 days on either side) and non-episodic days (remainder when BS <500 μg/m^3) separately and attempted to separate the effects of particles and SO_2. In discussing their regression analyses, the authors point out that "in the non-episodic period when the pollutants (SO_2 and BS) are considered separately, the coefficients are positive, significant, and similar. When they are considered together, only the coefficients for smoke remain statistically significant" (Mazumdar _et al._, 1981). This analysis provides clear support for the possibility of health risks associated with BS concentration below 500 μg/m^3 over 12 London winters, and suggest that SO_2 might be less important in non-episodic mortality. Because the early winters contained no non-episodic periods, the non-episodic analysis actually reflects only winters from 1960-61 to 1971-72, a time span that includes a significant change in the composition of London particulate matter as smoke controls were implemented. That the regression still indicates significance for the combined non-episodic days in these winters gives some confidence that the results may be relevant to areas with different particle composition, lower humidities and fewer periods of intense fog.

A comparison of the various analyses of London mortality (CD, Section 14.3.1.2, Appendix D) show qualitative similarities and suggest that smoke accounts for a small but statistically significant portion of daily mortality in London. The quantitative estimates vary with model and, as noted in the criteria document, the estimates should be viewed with caution and are quite uncertain, particularly at lower concentrations. No basis exists for selecting any of the approaches as most representative, although the Mazumdar analysis does reflect a much larger number of observations and conditions. Mazumdar et al. (1981) indicate linear and quadratic models are "both compatible with the data" and suggest consideration of controlled experiments or of further analyses.

Although the data are uncertain, the London mortality analyses suggest a risk of effects below the more certain effects levels of 500-1000 $\mu g/m^3$. Based on the comparison of analyses in the criteria document (CD, Section 14.3.1.2 Table 14-7), some small risk may exist at BS levels as low as 150 $\mu g/m^3$; the risk of any effect as well as the potential magnitude of the effect increases in the range of 150 to 500 $\mu g/m^3$. The criteria document (CD, p. 14-26) suggests that qualitative support for "weak but positive" associations between mortality and non-episodic particle concentrations (5-6 CoHs) is provided by analyses of New York City data by Schimmel and Murawski (1976), Schimmel (1978), and others (summarized in Table B-10). Qualitative data from regression analyses (Table B-11) are not inconsistent with the possibility of cumulative short-term mortality effects in U.S. atmospheres although their inherent difficulties limit their utility even in a qualitative context. Data from controlled human, animal, and other epidemiological studies, outlined in Section V.A. and B, suggest mechanisms by which various exposures to particles might result in mortality in susceptible individuals, but do not provide evidence for specific levels of concern.

Lawther's studies of bronchitic patients began during periods of high pollution (1954) and continued through the time when levels were considerably lower (e.g., 1967-68; mean winter SO_2, 204 $\mu g/m^3$, BS, 68 $\mu g/m^3$). Lawther et al. (1970) found an association between peak pollution and health status of bronchitics and that responses declined as controls reduced pollutant levels. Because of the nature of the study, effects were related to peak daily concentrations, but the authors felt the effects were more likely the result of brief exposures to short-term peaks "several-times the 24-hour average." While this suggestion is plausible, the available data preclude any quantitative evaluation. Thus 24-hour, or to a lesser extent, long-term pollutant averages, must be used to indicate the possibility of peak short-term levels of concern. Lawther's suggestion that 24 hr. averages of 250 $\mu g/m^3$ smoke and 500 $\mu g/m^3$ SO_2 represent "the minimum pollution leading to any significant response" appears reasonable, although the possibility that short-term peaks of concern may occur at lower 24-hour levels cannot be discounted. The criteria document notes that a summary of results for selected patients for the winter 1967-68 (Table III, Lawther _et al_., 1970) suggest a statistically significant correlation between smoke and symptom scores for a winter with only one day of $SO_2 > 500\ \mu g/m^3$ and BS $\geq 250\ \mu g/m^3$. The criteria document also suggests that BS measurements after 1963 may have overstated actual levels (CD, Table 14-7).

2. Translation to Thoracic Particle Indicator

A number of factors make use of these British studies in assessing risks of U.S. aerosols difficult. The most reliable conclusion that can be drawn is that as daily particle and SO_2 levels increase, mortality and symptoms increase. However, because the majority of U.S. studies of acute effects have been judged to be inadequate for quantitative evaluation of these effects, an attempt at translation of the British work must be made. The assessment outlined below incorporates reasonable assumptions concerning indicator pollutants, pollution composition, relative role of particles, and the nature of U.S. vs. British exposure regimes. In some cases assumptions are conservative (in the sense that they increase the margin of safety); in other cases their relative nature cannot be identified. This staff assessment is summarized in Table 6-2 and discussed below.

TABLE 6-2. STAFF ASSESSMENT OF SHORT-TERM EPIDEMIOLOGICAL STUDIES

	Measured British Smoke Levels (as $\mu g/m^3$)			Equivalent TP Levels ($\mu g/m^3$)
Effects/ Study	Daily Mortality in London[1]	Aggravation of Bronchitis[2]	Combined Range	Combined Range[3]
Effects Likely	500-1000	250*-500*	250-500	350-600
Effects Possible	150*-500	<250*	150-250	150-350

*Indicates levels used for upper or lower bound of range.

[1] Martin and Bradley (1960); Ware _et_ _al_. (1981); Mazumdar _et_ _al_. (1981)

[2] Lawther _et_ _al_. (1970)

[3] Boundary assumptions for estimating TP levels from BS readings detailed in text.

Both the mortality and bronchitic studies represent population groups among the most sensitive to pollutant effects. Because mortality, even in the sick and elderly population at greatest risk, is a more serious consequence than symptom aggravation, the risk of an effect at concentrations below the "effects likely levels" in the mortality studies should be given more weight than similar concerns for the bronchitic studies. As noted above and in Table 6-2, effects may be possible at the lower concentrations indicated, but the evidence and risks are much less certain. Therefore, the lowest pollutant levels of interest in the short-term studies were 150-500 $\mu g/m^3$ (BS) and 150-500 $\mu g/m^3$ (SO_2) (based on the mortality studies) and 250 $\mu g/m^3$ (BS) and 500 $\mu g/m^3$ SO_2 (based on the bronchitic studies). The relative importance of SO_2

cannot be unequivocally specified, but for reasons outlined in Section VI.A a separate particle standard is warranted. Thus, the conservative assumption (with respect to particles) is made that similar responses might have occurred without substantial amounts of SO_2 present. The Mazumdar _et al_. (1981) and Schimmel (1978) analyses suggest this might _not_ be a conservative assumption for mortality risks.

Because the smoke reading responds to darkness instead of mass, any relationship between BS and any mass index is particularly uncertain. Insofar as can be determined from the literature, smoke readings made prior to 1964 in British networks were not routinely calibrated against comparable mass readings (Holland _et al_., 1979). The most notable exception was the St. Bartholomew's site in central London (Waller, 1964). BS levels at this site are approximately related to particle mass less than about 5 μm. No such approximation is possible for the 7 stations in the exposure network used for the mortality and bronchitic studies, but the criteria document indicates that the central London site calibration tends to confirm "reasonably well" the mass calibrations in the 7 station network from 1959-63. After that time, BS readings may have overestimated actual concentrations (CD, Table 14-7). Comparisons of long-term averages of TSP vs. BS in central London from 1955 to 1963 suggest that when smoke levels in the range of 100-500 $\mu g/m^3$ are calibrated, they are consistently (about 100 $\mu g/m^3$) below TSP readings (Commins and Waller, 1967), further confirming the presence of particles larger than 5 μm, some of which must have been in the range that can be deposited in the thorax. At sites where BS is not calibrated to sampled mass (e.g. Salford in Lee _et al_. 1972), seasonal BS readings may actually _exceed_ TSP readings.

Although far from conclusive, the available data as discussed above and in Section IV suggest the following generalizations on the comparison between BS and TP (< 10 μm) are reasonable for the historical London data:

1) When London BS readings are in the range of 100-500 μg/m^3; fine particle mass < calibrated BS value < TP mass < TSP mass.
2) Where BS values are not calibrated, the uncertainty in these general relationships increases significantly; the extent of this uncertainty is difficult to assess.
3) Given the uncertainties in the uncalibrated data, it may be reasonable, but not highly conservative, to assume for historical (1959-72) London data, during the lower BS readings of interest (100-500 μg/m^3 BS), the following boundaries;
    a) As a lower bound, BS reading ≃ TP mass.
    b) As an upper bound, BS reading < TP mass ≃ TSP mass.
    c) As an upper bound for multiple daily readings, TP mass ≃ TSP mass = BS reading + 100 μg/m^3 (Holland *et al.*, 1979).

Based on these general boundary relationships, 24 hour TP levels of interest derived from the British studies would range between 150 μg/m^3 and 350 μg/m^3 (Table 6-2). By virtue of the assumptions in their derivation, these estimates should *not* be considered as demonstrated "effects levels."

Although no generally applicable relationships exist for predicting TP levels from specific BS readings, the available aerometric evidence suggests that the above ranges represent true boundaries that are likely to encompass the majority of values. In this regard, lower bounds derived from the above relationship include an additional margin of safety over that derived from the assessment of health data in the original units while upper bounds would tend to reduce any margin of safety incorporated in the original assessment of health data. Thus, under British conditions, the upper end of the range of interest (350 $\mu g/m^3$) overlaps levels at which effects are likely and does not include any margin of safety.

ii) Additional Factors to be Considered in Evaluating Margins of Safety and Risks - Short-term Exposures

As applied to the British exposures and particulate composition of 1959-1965, TP standards in the above range would provide a moderate to negligible "margin of safety" for the sensitive populations and effects studied. When applied to U.S. pollution, the following additional factors should be considered:

1. Aerosol Composition

1) Where high SO_2 levels are present with the above TP levels, the risks of effects are increased; any margin of safety inherent in applying the London data to U.S. areas with low SO_2 is reduced.

2) Where high photochemical smog levels are present, effects not accounted for by the British data may ensue.

3) Where U.S. particle components differ substantially from British particles, risks will vary.

As indicated in Section IV, it is difficult to compare acute toxicity of U.S. and British particles on a unit mass basis. The variability of TP composition among U.S. cities might be as great as that between any single U.S. city and historical London. Many U.S. urban areas have lower primary carbonaceous material, and higher sulfate, nitrate and secondary organics than London. This may not be the case in areas with heavy residential wood burning. Some areas have a relatively higher proportion of coarse mode material in the TP range. Therefore, risks associated with daily levels of 150-350 $\mu g/m^3$ TP will vary among U.S. cities, but it is difficult to predict the magnitude of the variability based solely on composition.

2. Relative Exposure

Based on measurement of indoor exposures and general ventilation rates reported by Lawther et al. (1970), indoor pollutant exposures in British residences more closely tracked outdoor levels than is the case in more tightly sealed U.S. residences. Thus, for comparable outdoor concentrations, the overall exposure to maximum 24-hour outdoor pollution was likely to have been greater in urban areas of Great Britian than in contemporary U.S. exposure situations. Since many of the more sensitive individuals may be confined indoors, the extent of increased mortality or symptoms would tend to be lower in the U.S. than observed at comparable levels in the British studies.

3. Risks for Other Sensitive Groups, Effects Not Evaluated

Based on the evaluation of toxicological, controlled human, and qualitative epidemiological data, Section V-B and Table 5-3 identify a number of groups that would be expected to be sensitive to ambient particles and a variety of effects that have been observed or anticipated to occur as a result of such exposures. The studies used to derive ranges address a number of these groups and effects, but omit others. Specifically, the derived range addresses 1) premature mortality in very sensitive individuals with chronic respiratory and cardiovascular diseases, individuals with influenza, and the elderly, and 2) morbidity (aggravation of disease) in bronchitic patients. Because it is reasonable to expect morbidity at or below levels at which mortality occurs in these sensitive groups, the London mortality studies also may be considered as an indicator of morbidity. Due to the unequivocal nature of the end point, the unusually large study population and substantial number of study days, the chance of detecting any marginal mortality effects is likely to be greater than for most studies of morbidity. Thus, to the extent the London data suggest a small risk of slight increases in mortality in the range of concern, an additional risk of morbidity is suggested and should be considered. Because the range is substantially below the mortality "effects" levels, the derived range would appear to be protective against the unmeasured morbidity risk.

Other sensitive groups not expressly addressed by the short-term British studies are children and asthmatics. Daily exposure to particles and SO_2 appear to be associated with increased symptoms of respiratory disease, particularly in sensitive children (Lebowitz et al., 1972). Other qualitative studies (see Table B-3) suggest possible effects of particulate matter on lung function in children and exercising adolescents in contemporary U.S. cities (Lebowitz et al., 1974; Dockery et al., 1981). The significance of these preliminary findings is not clear, however, and no reason exists to suggest important effects below the range noted above. Animal and controlled human exposures support the notion that acute exposure to a variety of particles can affect lung function (Section V.B.1) but usually only at high short-term (minutes to hours) exposures.

Asthmatics have proven difficult to evaluate in community studies. Clinical evidence suggest that they may respond to SO_2 alone (or in combination with particles) at levels likely seen in a number of community studies (e.g., Sheppard et al., 1981; Koenig et al., 1981). This confounds the results of studies where high SO_2 levels also were present. As discussed in the criteria document (CD, p. 14-30 to 14-33), both positive and negative findings have been reported for asthma and few studies have adequately controlled for temperature or other confounding variables. Thus, although clinical studies (Utell et al., 1981) and mechanistic considerations

(Boushey et al., 1980) suggest asthmatics are a sensitive group, the data do not support the existence of significant effects below the suggested range.

The above discussion addresses, in part, three of the six major categories of expected effects of particulate matter: 1) respiratory mechanics and symptoms; 2) aggravation of existing disease; and 3) mortality. The available data summarized in Appendix B suggest that two other categories (morphological alterations and carcinogenesis) may be better related to long-term exposure. Qualitative evidence summarized in Section V.B. suggests acute exposure to high levels of specific particles and community air pollution can affect clearance and other host defense mechanics, possibly resulting in increased infections and disease. In children, such infections might have longer-term consequences (Table 5-2).

Studies of hospital admissions or emergency room visits, although a crude indicator of morbidity, sometimes can provide some suggestion of the effects of pollution on several of the above categroies. The results of Samet et al. (1981), although essentially negative, provide a suggestion of a very small, but statistically significant, association between daily particle loadings and emergency room visits for respiratory diseases for TSP levels in the range of 14 - 700 $\mu g/m^3$ with SO_2 levels of 2 - 369 $\mu g/m^3$.

b) Long-Term Exposures

i) Derivation of Ranges of Interest from Epidemiological Studies

Cross-sectional and longitudinal studies most useful in establishing ranges of interest for long-term (annual) exposures for particulate matter are summarized in Section V-D. As noted in that section and in the criteria document, cross sectional studies are subject to a number of methodological uncertainties and confounding variables, such as differences in smoking, occupational exposures, and socioeconomic status among communities. Moreover, differences may exist in particle composition among areas, magnifying or diminishing apparent differences in effects. Nevertheless, the studies listed in Table 5-5, as well as more qualitative studies, suggest a relationship between various health responses and long-term exposures to particles.

A key feature of studies where more than two geographic areas are compared is the existence of a gradient of exposure and response for a number of health indicators. Examples suggesting a continuum of response from several qualitative European cross-sectional studies include Douglas and Waller (1966), Lunn et al. (1967) (see CD, Figure 14-4) and, less reliably, Lambert and Reid (1970). Although no obvious thresholds exist for many responses, not all of the health responses measured in these studies show a gradient.

When only two geographic areas or time periods are involved in a study, the levels that happened to exist in the more polluted area or time period usually are listed as the "effects" levels (Ware et al., 1981; CD, p. 14-49). Effects levels thus derived are somewhat arbitrary, but do represent concentrations where effects can be ascribed with some certainty. Because a gradient of effects is often observed for multiple area studies, however, some risk exists that effects decrease but do not necessarily disappear at concentrations substantially below those of the more polluted area. Therefore, "effects" levels derived from two-area (or time) studies cannot be regarded as thresholds or "no detectable effect" levels.

Table 6-3 is a staff assessment of the levels of interest derived from the most useful long-term epidemiological studies. In this Table, the "effects likely" line lists concentrations in the more polluted area

TABLE 6-3. STAFF ASSESSMENT OF LONG-TERM EPIDEMIOLOGICAL STUDIES

	Measured BS Levels (as μg/m^3)	Measured TSP Levels			Equivalent TP Levels (μg/m^3)
Study/Effect	Increased Respiratory Disease, Reduced Lung Function in Children[1]	Increased Respiratory Disease Symptoms, Small Reduction in Lung Function in Adults[2]	Increased Respiratory Symptoms in Adults[3]	Combined Range	Combined Range[4].
Effects Likely	230-300 BS	180*	--	≥ 180	90-110
Effects Possible	< 230 DS	130-180*	60-150 (110*)	110-180	55-110
No Significant Effects Noted	--	80*-130	--	80-110	40-55

*Indicates levels used for upper or lower bound of range.

[1] Study conducted in Sheffield, England (Lunn et al., 1967).

[2] Studies conducted in Berlin, NH (Ferris et al., 1973, 1976).

[3] Study conducted in Ansonia, CT (Bouhuys et al., 1973)

[4] Conversion assumes range of TP/TSP of 0.5 to 0.6. See text.

(or time) as derived from the similar "effects likely levels" column of Table 5-5. For the Lunn study, effects are possible at lower concentrations, but lower boundaries cannot be specified. Based on the evaluation of the Lunn follow-up study (Lunn et al., 1970) in the criteria document, the "no effects" level for smoke sometimes ascribed to these studies (140 $\mu g/m^3$ BS; Holland et al., 1979) is not reliable. The "effects possible" levels for Ferris represent a lower bound for detectable effects in the Ferris studies, based on the lack of measurable differences in effects between years when TSP dropped from 130 to 80 $\mu g/m^3$. Based on the mixed but suggestive results from Bouhuys et al. (1978), effects are possible at levels observed in the more polluted city. Effects, if any, are most likely to have resulted from higher exposures in previous years (60 - 150 $\mu g/m^3$), with the median concentration (110 $\mu g/m^3$) serving as a reasonable lower bound.

Based on the above assessment, levels of interest for effects measured in these studies are as follows: 1) Decreased lung function and increased acute respiratory disease in children may occur at levels below 230 $\mu g/m^3$ BS (Lunn) 2) Decreased lung function in adults may occur at TSP levels as low as 130-180 $\mu g/m^3$ TSP (Ferris) but is less likely to be detectable at about 110 $\mu g/m^3$ (Bouhuys). 3) Some risk of increased respiratory disease and/or symptoms in adults may exist at levels of 110-180 $\mu g/m$ TSP (Bouhuys, Ferris).

It must be stressed that effects are not demonstrated within these ranges; the lower bounds represent conservative estimates where some risk of effect is not ruled out by the data. Moreover, the differences in effects at lower levels (e.g., Bouhuys) were, as might be expected, small in comparison to baseline rates.

Conversion of the BS data to TP is quite uncertain for these long-term studies because sites were located in different geographic areas and times and because no mass calibrations are available for these areas. Because reliable lower bounds cannot be estimated for the Lunn study and because the levels in the more polluted areas are relatively high, no attempt at translation of these results to thoracic particle equivalents will be attempted.

Approximate conversions of TSP data to that fraction more likely to have produced the observed responses (TP) may be accomplished by using relationships developed from reliable particle size distribution measurements and compared with data from the inhalable particle (IP) network (Watson _et al._, 1981). For TP <10 μm, the TP to TSP ratio expected for typical urban U.S. sites averages between 0.5-0.6. Although these average ratios might not have been obtained in the Ferris and Bouhuys studies, the range is probably reasonable and conservative. Applying these factors to the U.S. studies summarized above (Table 6-3), the range of some interest for TP is 55 to 110 $\mu g/m^3$. The upper end of this range overlaps the somewhat uncertain "effects level" derived from these studies. As such, the upper bound contains no identifiable margin of safety. It is emphasized that the results of the original studies, assessment of risks at lower levels, and conversion to a common indicator all are subject to considerable uncertainties.

ii) Additional Factors to be Considered in Evaluating Margins of Safety and Risks - Long Term Exposures

It is apparent that long-term concentrations in the range of 55-110 $\mu g/m^3$ are in the "noise level" for the above epidemiological studies and that effects measured to date are likely to be small for the groups studied. Preliminary results of cross sectional data from the ongoing "Six Cities Study" tend to support this judgment (Ferris et al., 1979, 1980). When evaluating margins of safety (risks) in this range, the following additional factors should be considered.

1. Aerosol Composition:
    1) Because SO_2 levels in New Hampshire were generally low, the Ferris study provides some support for separate standards. Where high SO_2 levels co-exist with TP, however, the lower portion of the above range still is protective.
    2) The Bouhuys and perhaps the Ferris studies are directly relevant to current U.S. atmospheres with periodic elevations of ozone.
    3) Although relative aerosol toxicity among the various regions studied cannot be compared on a unit mass basis, the results of the above analysis of U.S. and British studies do not suggest an order of magnitude difference. Each area, however, probably contained city specific sources producing somewhat different particle composition. For example, particles in the city studied by Ferris were largely affected by pulp mill emissions. As such, their composition may be somewhat unusual, but no measurements are available. The risks of lung function and symptomatic effects noted in these chronic studies can be expected to vary with composition, but reliable estimates of the variability are not available.

2. Relative Exposures

The Bouhuys and possibly the Ferris studies may represent contemporary U.S. indoor/outdoor exposures. It is apparent, however, that at lower outdoor particle levels ($< 130\ \mu g/m^3$ TSP), longer term particle exposures may be significantly affected by indoor pollution. With one or two smokers present, indoor particle exposures will exceed outdoor exposures. This can be viewed in two ways: a) reduction in outdoor particle levels will not produce proportional reduction in total exposures; or b) in terms of total body burden, the uncontrolled indoor contribution reduces the subject's capacity for dealing with outdoor levels, thereby increasing concern over both sources of exposure. About 50% of homes, however, have no smokers in them and indoor particle generation is normally much smaller (Spengler *et al.*, 1980).

3. Risks for Other Sensitive Groups, Effects Not Evaluated

The groups examined in the studies used to derive ranges include children and adults in the general population. Effects categories addressed include 1) respiratory mechanics and symptoms and 2) effects on host defense mechanisms, as suggested by increased respiratory diseases. Different effects categories and other, more sensitive, subgroups were not analyzed separately. Although children can, on the basis of mechanistic and activity pattern arguments, be considered a sensitive group, they are not necessarily the most sensitive.

Other sensitive groups listed in Table 5-3 not directly addressed by the long-term studies include asthmatics, bronchitic subjects, the elderly, and individuals with cardiopulmonary disease. Based on the summary of mechanisms and qualitative effects data in Section V, these groups would appear most likely to respond to single or repeated acute exposures. They should be largely protected against such effects by the 24 hour standard. However, these as well as other sensitive groups in Table 5-3 may be subject to additional potential effects of concern associated with long-term exposures that were not adequately addressed in the studies used to derive ranges. These potential effects include:

1) Direct or indirect effects on lung tissues contributing to development or aggravation of emphysema, cardiopulmonary disease, and pneumoconiosis. Controlled animal and human studies of the effects of acid aerosols on clearance (Table B-5), community studies of high long-term exposures to sulfur oxide-particle pollution (Table B-7), the occurrence of industrial bronchitis with very high mineral dust exposure (Morgan, 1978), and the well established effects of cigarette smoke suggest that long-term exposures to repeated peaks of ambient particles, particularly acid aerosols, may play a role in the etiology of chronic bronchitis, and ultimately aggravation of emphysema and associated cardiopulmonary difficulties. Toxicologic studies of high exposures to sulfuric acid/ammonium sulfate SO_2 aerosols (Table B-8) and autopsies of humans and zoo animals (Table B-9) show that long-term exposures to both acid aerosols and crustal dusts can damage or alter lung tissues. Acid aerosols effects were related in one study to an incipient stage of emphysema, while crustal dusts produce silicate pneumoconioses with varying degrees of clinical significance. Although these qualitative data generally involve exposures higher than any expected with the derived range, the possibility of some risks at lower exposures should be considered.

2) Carcinogenesis - Cigarette smoking generally is recognized as the major determinant of lung cancer. The available epidemiological evidence

does not unequivocally show that current particle exposures contribute to cancer nor does it rule out the possibility of some small contribution. The presence of mutagens in organic particulate fractions from unidentified sources and the potential interaction between these, as well as inert particles, and carcinogens from cigarettes or occupational exposures (Appendix B, Section E), suggest some need for caution and further study.

3) Premature Mortality - Regression analyses of long-term mortality (Table B-11) have been used to suggest effects of sulfur containing particulate pollution. The criteria document discusses a number of inherent difficulties and inconsistencies that preclude quantitative conclusions and limit their qualitative use.

The available information does not suggest major risks for these effect categories at current ambient particle levels in most U.S. areas. Nevertheless, the risk that both fine and coarse mode particles may produce these responses supports the need to limit long-term levels of TP for a variety of aerosol compositions.

D. Summary of Staff Conclusions and Recommendations

The major staff conclusions and recommendations made in Section VI. A-C above are briefly summarized below:

1. A separate general particulate standard remains a reasonable public health policy choice.
2. The current TSP standard directs control efforts towards particles of lower risk to health because of its inclusion of larger particles which can dominate the measured mass concentration, but which are deposited only in the extrathoracic region. A new particle indicator representing those particles capable of penetrating the thoracic regions (thoracic particles, TP)

is recommended. The size range should include those particles less than a nominal 10 μm and sampler performance criteria should be related to respiratory tract deposition data.

3. Both short-term (24-hour) and annual arithmetic mean standards are recommended. The short term standard should be expressed in statistical form, the decision on allowable number of exceedances should be made in conjunction with establishing a level for the standard.

4. Based on a staff assessment of the short-term epidemiological data, the range of 24-hour TP levels of interest are 150 to 350 $\mu g/m^3$. Under the conditions prevailing during the London studies, the upper end of the range represents levels at which effects are likely in the sensitive populations studied. Given the uncertainties in translating these results to U.S. conditions and the seriousness of the potential health effects, the upper end of the above range contains no identifiable margin of safety and should not be considered as an appropriate standard alternative. The uncertainties and the nature of the potential effects are important margin-of-safety considerations. Neither the studies used to derive the range nor more qualitative studies of effects in other sensitive population groups (e.g. asthmatics, children), or effects in controlled human or animal studies provide scientific support for health risks of consequence below 150 $\mu g/m^3$. These qualitative data as well as factors such as aerosol composition and exposure characteristics should also be considered in evaluating margins of safety associated with alternative standards in the range of 150 $\mu g/m^3$ to something below 350 $\mu g/m^3$.

5. Based on a staff assessment of the long-term epidemiological data, the range of annual TP levels of interest are 55 to 110 $\mu g/m^3$. The upper end of this range overlaps the somewhat uncertain "effects levels" derived from these studies. Due to these uncertainties, the upper end of the range (110 $\mu g/m^3$) may not include any margin of safety, and should not be considered as an appropriate standard alternative. The lower end (55 $\mu g/m^3$) represents a level where some risk of symptomatic effects might remain but no detectable differences in pulmonary function or marked increases in respiratory diseases are expected. Increases in symptomatic effects at the lower levels are uncertain and small in comparison to baseline rates.

When evaluating margins of safety for an annual standard, it is particularly important to examine the results of qualitative data from a number of epidemiological, animal, and air quality studies. These suggest concern for effects not directly evaluated in the studies used to develop the ranges. Such effects include damage to lung tissues contributing to chronic respiratory disease, cancer, and premature mortality. The available scientific data do not suggest major risks for these effects categories at current ambient particle levels in most U.S. areas. Nevertheless, the risk that both fine and coarse particles may produce these responses supports the need to limit long-term levels of TP for a variety of aerosol compositions.

Because of different form, averaging procedures and size range, precise comparisons between the above ranges of TP standards and the current primary TSP standards are not possible.* The lower bounds, taken together, result in standards roughly equivalent in stringency to the current standards. In general, the rest of the ranges represent varying degrees of relaxation as compared with the current standards. At the lower concentrations in the ranges much of the relaxation would result because only smaller particle sizes would be collected. Thus, a city where exceedance of the TSP standard was largely dominated by coarse mode dust (with substantial mass of particles greater than 10 μm) would be less likely to violate a comparable TP standard than would an area where exceedance of the TSP standard was dominated by particles smaller than 10 μm. At higher concentrations in the above ranges, standards would permit increased levels for TP as well as for larger particles.

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*By applying observed TP/TSP ratios and other factors, crude comparisons can be made. The current annual TSP standard (75 μg/m^3, geometric mean) is roughly equivalent to an arithmetic mean of 50 μg/m^3 as TP. The numerical value of the 24 hour TSP standard (260 μg/m^3) is roughly equivalent to 140 μg/m^3 TP, but this does not account for differences between the arithmetic (current standard) and recommended statistical form.

VII. CRITICAL ELEMENTS IN THE REVIEW OF THE SECONDARY STANDARD

This section discusses information drawn from the criteria document that appears most relevant in the review and possible revision of secondary standards for particulate matter. Four major categories of welfare effects are examined. Within each category, the paper presents 1) a brief summary of relevant scientific information, 2) an evaluation of potential quantitative relationships between atmospheric particles and effects, and 3) a staff assessment of whether the available information suggests consideration of secondary standards that differ from the recommended primary standards. Where applicable, preliminary staff recommendations are presented on the most appropriate pollutant indicator and form of standards, in the event the Administrator determines such standards are warranted.

A. Visibility and Climate

1. Nature of Effects

a) Visibility

i) Categories and Extent of Perceived Effects

Impairment of visibility is perhaps the most noticeable and best documented effect of particles in current U.S. atmospheres. Although often equated with "visual range" as measured by airport weather observers, visibility in a broader context relates to visual perception of the environment and involves color and contrast of viewed objects and sky, atmospheric clarity, and the psychophysics of the eye-brain system (CD, p. 9-43 to 9-46).

For present purposes, it is useful to classify pollution-derived effects on visibility into two categories: 1) regional haze and 2) visible plumes. The nature and extent of these effects are determined largely by the distribution and characteristics of anthropogenic and natural particles and, to a lesser extent, by NO_2. Salient features of both categories are outlined below.

Regional haze is relatively homogeneous, reduces visibility in every direction from the observer, and can occur on a geographic scale ranging from an urban area to multistate regions. Increased haze reduces contrast causing more distant objects to disappear. Nearby objects can appear "flattened" and discolored, the horizon sky is whitened, and scattered light is perceived as a grey or brown haze (Charlson _et_ _al_., 1978). When urban light and haze combine at night, the contrast between the night sky and the stars is reduced, markedly limiting the number of stars visible in the night sky (Leonard _et_ _al_., 1977).

Visual range (visibility) measurements at airports are a useful indicator of the extent and intensity of regional haze. Analyses of airport visibility trends from 1948 to 1972 suggest that while visibility in some urban areas improved or stayed the same during the winter months, the occurrence of episodic eastern U.S. regional haze with visibilities less than seven miles appears to have increased. This later occurrence has been most notable during the summer months (Husar _et_ _al_., 1980). Since 1972, regional visibility in both the East and West apparently has improved slightly but not to pre-1960 levels (Sloane, 1980; Marians and Trijonis, 1979). Whether this recent improvement is related to more favorable meteorology or reduced regional particle and sulfur oxide emissions is not known with certainty, but such reductions are reflected in emissions inventories in both East and West. Regional differences in visibility are illustrated in Figures 7-1a and 7-1b. As indicated by suburban and non-urban airport data, visibilities in the East are substantially

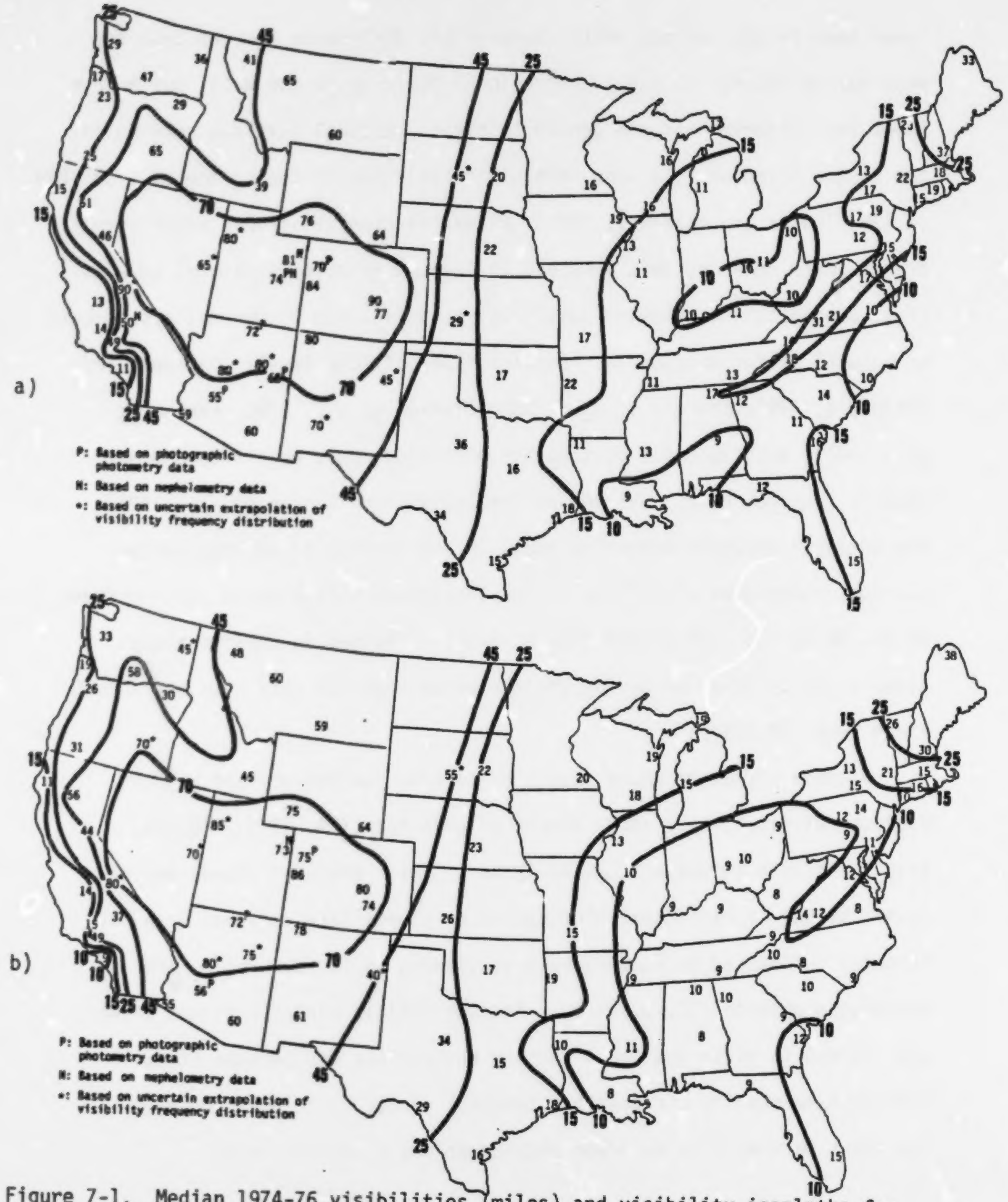
a)
P: Based on photographic photometry data
N: Based on nephelometry data
*: Based on uncertain extrapolation of visibility frequency distribution
b)
P: Based on photographic photometry data
N: Based on nephelometry data
*: Based on uncertain extrapolation of visibility frequency distribution
70
45
25
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Figure 7-1. Median 1974-76 visibilities (miles) and visibility isopleths for suburban/nonurban airports: a) yearly and b) summertime (Trijonis and Shapland, 1979). Data are subject to uncertainties associated with suburban airport observations, but show general regional patterns. The clear differences between East and West are parallelled by regional humidity (CD, Figure 1-17) and nonurban fine particle levels. Summertime fine mass averaged from 22 to 25 $\mu g/m^3$ at 12 eastern nonurban sites (Watson *et al.*, 1981) and about 4 $\mu g/m^3$ for 40 Rocky Mountain and southwest background sites (Snelling, 1981).

lower than in most of the West. Some of the difference between East and West may be related to the lower regional humidity in the West, but a more important difference is the generally higher regional particle loading in the East. Based on: 1) long-term historical data in the northeast from 1889 to 1950 (Husar and Holloway, 1981); 2) examination of airport visibility trends after deleting data possibly influenced by obvious natural sources (fog, precipitation, blowing dust) (CD, p. 9-75); and 3) current assessments of natural sulfur sources and regional fine particle levels (Galloway and Whelpdale, 1980; Stevens _et al_., 1980; Pierson _et al_., 1980, Ferman _et al_., 1981), anthropogenic particulate pollution would appear to dominate eastern regional haze. Relying on the analysis of Ferman _et al_. (1981), the criteria document estimates that, in the absence of anthropogenic sources, summertime visibility in the Shenandoah Valley would range between 60 and 80 km (36 - 50 miles) (CD, p. 9-63). The median daytime visual range actually observed during the one month study at this site was 4 to 5 times lower (9 miles).

Visible plumes of smoke, dust, or colored gas obscure the sky or horizon relatively near their source of emission (EPA, 1979). Black, gray, or bluish plumes are caused by particles. Brownish plumes may be caused by NO_2 or particles. Perception of plumes (and regional haze) is strongly influenced by factors such as viewing angle, sun angle, and background objects (CD, p. 9-46). Because visible particle plumes often are subject to state and local opacity regulations and because it is difficult to specify air quality standards to deal with elevated plumes, the focus here will be on urban and larger scale regional haze.

ii) Evaluation of Visibility

Visibility impairment may adversely affect public welfare in essentially two areas: 1) the subjective enjoyment of the environment (aesthetics, personal comfort and well-being); and 2) transportation operations. As discussed in the criteria document, evidence on the effects is drawn from studies of social perception and awareness of air pollution, economic studies, and visibility/air transportation requirements. These studies are discussed and evaluated in Appendix C. Key findings are summarized in Table 7-1 and discussed below.

1) Early studies of social awareness (e.g., Schusky, 1966) have found that as particulate pollution levels increase, an increasing portion of the population is aware of air pollution and considers it a nuisance. A more recent Los Angeles study (Flachsbart and Phillips, 1980) found that of nine pollutant indices, only two, visibility and ozone, were consistently and significantly related to perceived air quality for all averaging times.

2) Economic approaches to evaluation of visibility include "iterative bidding" and property value studies. Despite their limitations for providing quantitative estimates, iterative bidding studies (see Table C-1) suggest that visibility is of substantial economic value in both urban and natural settings. Although the value of visibility in other areas may vary significantly from that suggested for the areas studied to date (chiefly the rural southwest and Los Angeles), no *a priori* reason exists to suggest that visibility is of little value in heavily populated eastern urban areas or in widely visited eastern recreational areas. The Los Angeles iterative bidding study (Brookshire *et al.*, 1979) suggests that about 2/3 of the estimated willingness

TABLE 7-1. SUMMARY OF QUALITATIVE EVIDENCE FOR VISIBILITY RELATED VALUES

	Effect of Increased Visibility	Affected Groups	Averaging Times*	Supporting Observation
Transportation	More efficient, lower risk operations, visual approach permitted	Airport users, operators civilian and military	1-3 hr. readings	Visual approaches permitted when visibility >3-5 miles; airport specific (FAA, 1980b)
	Increased opportunity to operate aircraft	General aviation aircraft (non-instrument capable pilots, aircraft)	1-3 hr. readings	VFR permitted when visibility >3 miles (FAA, 1980a)
Aesthetic: 1) Social Criteria	Decreased perception of air pollution	Substantial percentage of general population; urban areas	Daily to annual	Perception of air pollution in Los Angeles significantly related to visibility for all averaging times (Flachbert and Phillips, 1980). Perception, annoyance significantly related to particulate matter (Schusky, 1966)
	Options values; maintaining or increasing opportunity to visit less impaired natural and urban settings	Outdoor recreationists, campers, tourists	Daily, peak visitation in summer months	Aggregate of activity values in iterative bidding studies suggests importance of options values (Rowe and Chestnut, 1981)
	Improved view of night sky	Amateur astronomers, other star watchers	Nightly	Decrease in star brightness by fine particles (Leonard et al., 1977)
2) Economic Criteria	Increased property values	Home owners	Long-term	Property values related to perception of air pollution, hence visibility (Rowe and Chestnut, 1981; Brookshire et al. 1979)
	Enhanced enjoyment (user or activity values) of environment in:			
	a) Urban settings	Urban dwellers	Long-term	Willingness to pay for increased visibility in urban (Brookshire et al., 1979) and non-urban settings (Rowe et al., 1980)
	b) Natural settings	Outdoor recreationists, campers, residents of non-urban areas	Daily, peak visitation in summer Months	
3) Psychological Criteria	Existance values; maintaining pristine environments	General population	Long-term	Existance values may far outweigh activity or user values (Rowe and Chestnut, 1981)
	Less concern over perceived health effects	General population, urban areas	Daily to Long-term	About 2/3 of bid for improved visibility in Los Angeles was related to concern over potential health effects (Brookshire et al., 1979)

*Represents staff judgment of most important averaging time based on supporting observation. Because averaging times are related it is difficult to specify single (long or short) averaging time as most significant. Perception of visibility is essentially instantaneous.

to pay for visibility improvement was related to concern over potential health effects.

3) Property value studies are less useful indicators of visibility values, but when combined with perception and bidding studies, the data suggest that visibility may have tangible effects on property values in urban areas.

4) The available information indicate that the major effects of visibility on transportation are related to air traffic, particularly when visibility falls below three miles and FAA restricts visual flight (VFR) operations. The data show that episodic regional haze over large segments of the East tends to curtail some segments of general aviation aircraft and slow commercial, military, and other instrument flight (IFR) operations on the order of 2-12% of the time during the summer. The extent of any delays varies with airport. Reduced visibility also may tend to increase risks associated with aircraft operations in the mixing layer, but quantitative assessments are not available.

b) Climate

The presence of particles that scatter and absorb light also may affect climate. Principal effects of concern include:

1) Reduction of net solar radiation to the earth's surface--this may lead to small local or regional reductions in temperature and ultimately affect climate. Trends in atmospheric turbidity (a measure of the reduction in the intensity of direct sunlight reaching the ground) are qualitatively similar to those for visibility (CD, Figure 9-37).

2) Enhanced cloud and fog formation, and possible increased precipitation (CD, p. 9-95). Enhanced fog formation would add to the adverse visibility effects discussed above.

Because available information as summarized in the criteria document does not permit quantitative assessment of the relationship between the presence of particles and effects on climate, such effects, although potentially of significance, cannot form the principal basis for a secondary particle standard at this time. Nevertheless, because effects of particles on climate and on visibility both appear to be dominated by fine particles, the potential climatic effects would tend to provide support for decisions based on visibility criteria.

2. Mechanisms and Quantitative Relationships

The more important studies in the criteria document on mechanisms by which particles impair visibility and quantitative relationships between particles and visibility are evaluated and discussed in Appendix C. Key conclusions are summarized below.

1) Visibility impairment is caused by light scattering and absorption by atmospheric particles and gases. Theoretical and empirical results provide strong evidence that visual range reduction in urban and regional haze normally is controlled by fine particles (<2.5 μm). The only important situations where larger particles dominate are some naturally occurring phenomena including precipitation, fog, and dust storms.

2) The relative importance of scattering and absorption by particles and the extinction (scattering and absorption) efficiency per unit mass of fine particles vary with chemical composition and, to some extent, humidity. Scattering dominates regional haze, but absorption can be important in urban settings. As humidity increases from 70 to 90%, fine particle scattering can increase by a factor of about two.

3) Two fine particle components, hygroscopic sulfates and elemental carbon, generally tend to be the most important in reducing visibility. Sulfate compounds with associated water often dominate fine mass and light scattering, particularly in the East, while elemental carbon accounts for most particle absorption in urban areas.

4) Within reasonable limits, the Koschmieder equation (Equation C-1) can be used to relate visual range and fine particle concentration. To do this, the Koschmieder constant (K) must be estimated. Available data (CD, p. 9-9) suggest that due to imperfect techniques and observers, reported K varies from the commonly used value of 3.9 (ranging from 1.7 to 3.6). Figure 7-2 illustrates the Koschmieder equation for the standard $K = 3.9$ and a representative range of fine particle extinction efficiency per unit mass (γ). Cases are detailed in Appendix C.

3. Staff Recommendations

a) Rationale for Consideration of a Secondary Standard

Impairment of visibility over urban to multistate regions is clearly an effect on public welfare as specified in the Clean Air Act. The relationship of fine particles to visibility impairment in such cases is known well enough to permit quantitative predictions of the ranges of visibilities for given fine particle levels. Of initial concern is whether the range of recommended primary standards would provide adequate protection against these welfare effects. Rough estimates of visibilities associated with alternative primary standards are listed in Table 7-2.

Precise comparisons of predicted visibility associated with primary standard attainment with current airport visibility information are difficult. It would appear, however, that none of the alternative

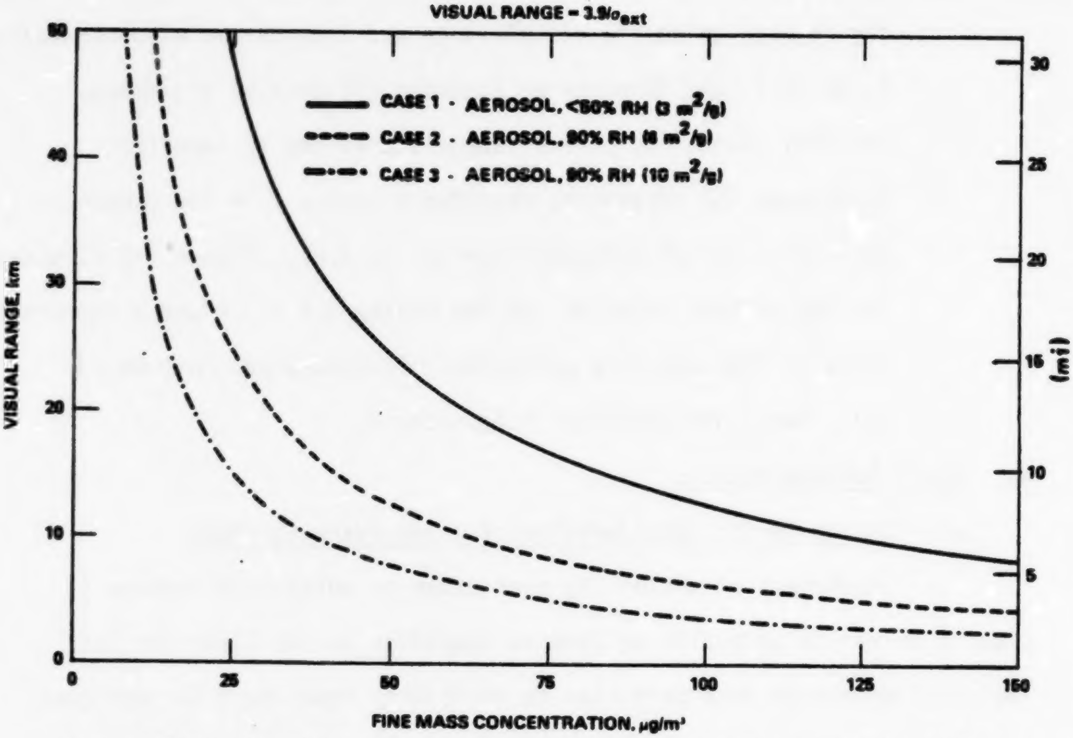

Figure 7-2. Visual range as a function of fine mass concentration (determined from equilibrated filter) and γ, assuming the "standard" K = 3.9. Because K is commonly lower in non-ideal application, results from this relationship should not be compared directly to airport visibility data.

TABLE 7-2. VISIBILITY ASSOCIATED WITH ALTERNATIVE PRIMARY STANDARDS

TP (< 10 μm) Standard		Range of Visibility (miles)*
24-Hour	150	1.9 - 6.2
	250	1.1 - 3.8
	300	1.0 - 3.1
Annual	55	8.4 - 16
	75	6.2 - 12
	100	5.2 - 10

*Range derived from application of relationship in Figure 7-2 with K = 3.9 and γ = 4 to 8 m^2/g. Comparable visibilities reported by airports would be lower. For 24 hour cases fine mass/TP mass ratios are assumed to range from 0.6 - 1. For annual cases the ratio was fixed at 0.6. Primary standards are assumed to be just met throughout the viewing distance. Because primary standards usually are directed at maximum point measurements, this assumption tends to understate visibility resulting from primary standard attainment. Nevertheless, it is not clear whether control strategies directed at attaining TP standards would reduce the fine fraction responsible for visibility impairment by the same proportion as total TP mass is reduced. The effect of 24-hour standard on annual visibilities was not evaluated. The annual values are made less certain because natural impairment (fog, rain) is not included and because the arithmetic mean particle mass was used to derive visibility. These errors tend to be in opposite directions. The visibility estimates for the annual standards are, therefore, best interpreted as representative of "typical" days rather than as annual averages.

primary standards would materially improve regional visibility over current levels unless attainment strategies included regional fine particle controls. With the exception of the lower half of 24 hour values, most of these alternative standards might permit some decrease in visibility, even in the East.

The question then arises as to whether a separate secondary standard is needed to limit further impairment of visibility or improve visibility over levels permitted by the primary standards and other applicable regulations. Unfortunately, reliable quantitative information on the value of visibility on a national level is not available. Taken together, however, the qualitative benefits of improving or maintaining visibility summarized in Table 7-1 suggest that visibility values, although of uncertain magnitude, are important in a variety of contexts. Because of this, and the uncertainty as to whether the recommended primary standards would provide adequate protection of public welfare with respect to these values, the staff recommends that a visibility-related particle standard be considered.

b) Factors to Be Considered in Selecting a Standard

In the event the Administrator decides to propose a secondary standard, decisions will be necessary on the most appropriate pollutant indicator, averaging time, form, and level of the standard. Preliminary staff recommendations in each of these areas follow.

i) Pollutant Indicator

The indicator for the current secondary particle standard is TSP. The available information shows that TSP is a poor indicator for particles that impair visibility. The current secondary standard tends to focus controls on coarse particles with little resulting benefit for visibility. Alternative indicators for a visibility related secondary standard include:

1) Direct measurements of visibility and long-path extinction through
   - a) human observers/photography
   - b) telephotometers
   - c) transmissometers
2) Measurement of the optical properties of the aerosol
   - a) Light scattering (nephelometer)
   - b) Light absorption (carbon analysis or transmittance)
3) Measurement of fine particles
   - a) Fine mass
   - b) Specific chemical components

Direct measurement of visibility is attractive in that it is directly related to the effect, responds to changes in humidity (unlike heated nephelometers), and provides long-path integration of pollutant impacts. However, such measurements are labor-intensive, vary with observer, target and atmospheric conditions, and are affected by natural phenomena such as dust and fog as well as NO_2. These limitations would make specifying reference methods incorporating such approaches in support of particle-based air quality standards difficult. Thus, direct visibility measurements are not recommended as the principal indicator for standard compliance.

Measurement of the optical properties of the aerosol has a number of advantages. Nephelometers are available and provide continuous readings of light scattering by particles. The high correlation between fine mass and light scattering suggests the nephelometer is a good indicator of fine mass, and one that is somewhat sensitive to changes in composition that tend to affect scattering. Although routine measurements for light absorption are not yet available, promising methods are under development.

In the case of regional haze, however, absorption is generally small and measurements may be unnecessary.

Despite the advantages of nephelometers, it might not be wise to recommend these as the principal measurement method at this time. To date, they have been used principally by trained researchers. Large-scale networks with routine quality assurance programs have not been tested. Moreover, companion measurements of fine mass and composition probably would be needed for source assessment. If, however, short-term (<24 hour) average times become important, measurement of scattering by automated nephelometer might be the most appropriate particle indicator.

Although fine particle mass measurements are less directly related to visibility than scattering on an absolute basis, the two measurements are highly correlated at most sites. Furthermore, the relationship between fine particle mass and visibility is relatively straightforward. Fine particle instruments are available and long-term (1 year) data exist from at least three major networks (Pace et al., 1981; Cahill et al., 1981; Watson et al., 1981). Compositional analyses of mass measurements provide important insights as to the sources of the haze. Some obvious natural sources (coarse dust, fog) are excluded by such measurements.

Because sulfates and carbon are often the most significant fine particle components contributing to extinction, some consideration might be given to their use as standard indicators. A sulfate standard would direct control towards eastern regional visibility impairment, but have little impact in most western urban areas. Carbon standards would direct control towards urban areas, with little regional impact. Neither indicator would include other fine particles. If all fine particles are to be accounted for, specific substance indicators might be unnecessarily complex.

Considering the practical constraints associated with large monitoring networks and because fine mass may be better related to other welfare effects of fine particles (e.g., soiling, materials damage), than optical measurements, the staff recommends fine mass as the principal pollutant indicator. On both practical and empirical grounds, the commonly used D_{50} of 2.5 μm for fine mass is acceptable. Fine mass measurements could, in some cases, be supplemented by "equivalent" extinction measurements.

Because fine mass (and light scattering) instruments provide point measurements, it is recommended that concentrations from several instruments (three or more) be spatially averaged to provide a more reliable indication of the long-path concentrations that are more directly related to visibility. Separation of instruments should be on the scale of the desired visual range.

Spatial averaging tends to minimize the chance of singling out point source or urban "hot spots" of maximum concentration that are unimportant on a regional scale. Addressing regional scale visibility problems will require a fundamentally different control approach than has been used in the past for TSP. Available information on composition, extinction, and transport suggests that multistate regional scale control of the gas phase precursors (notably SO_2) of secondary aerosols would be considerably more important than additional controls on traditional local primary particle emitters (Lodge *et al*., 1981). A spatially averaged pollutant indicator should facilitate focusing on the more important sources.

ii) Averaging Times

As indicated in Table 7-1, visibility impairment can be of importance for hourly to annual (or longer) time scales. Although close control of appropriate and practical short- and long-term averaging times is important in establishing standards for protecting public health with an adequate margin of safety, it is less clear that multiple averaging times are necessary for welfare effects such as visibility.

For control purposes, longer averaging times are desirable in that effects of extreme weather and source conditions tend to be dampened. To the extent that long-term average visibilities correlate with the occurrance of the best and worst daily visibilities, a single long-term standard may be sufficient. This indeed seems to be the case. The evaluation of eastern airport visibility trends by Trijonis et al. (1978a,b) and Trijonis and Shapland (1979) indicates similar trends in best tenth percentile, worst tenth percentile, and median visibility. Figure 7-3 shows representative cumulative frequency plots for visibility. This nearly linear relationship is found at most Eastern and Western sites evaluated by Trijonis and co-workers (1978a,b). It thus appears that a single averaging time for visibility should provide a reasonable index for all averaging times. Spatially averaged fine particle levels may follow different, but still predictable, distributions. Thus, if the rationale for the standard were derived largely from a short-term effect (e.g., airport operation) a longer term standard could be specified to provide adequate protection within predictable statistical limits.

A seasonal averaging time may represent the most reasonable indicator of both long-term and short-term visibility values. Additional supporting factors include: 1) visibility in the eastern U.S. has a strong seasonal character; 2) the greatest impacts on recreation and transportation occur in the summer months; and 3) regional models currently under development can deal with seasonal averages better than with daily averages. The staff therefore recommends a calendar quarterly (three month) averaging time for a secondary visibility based standard. The

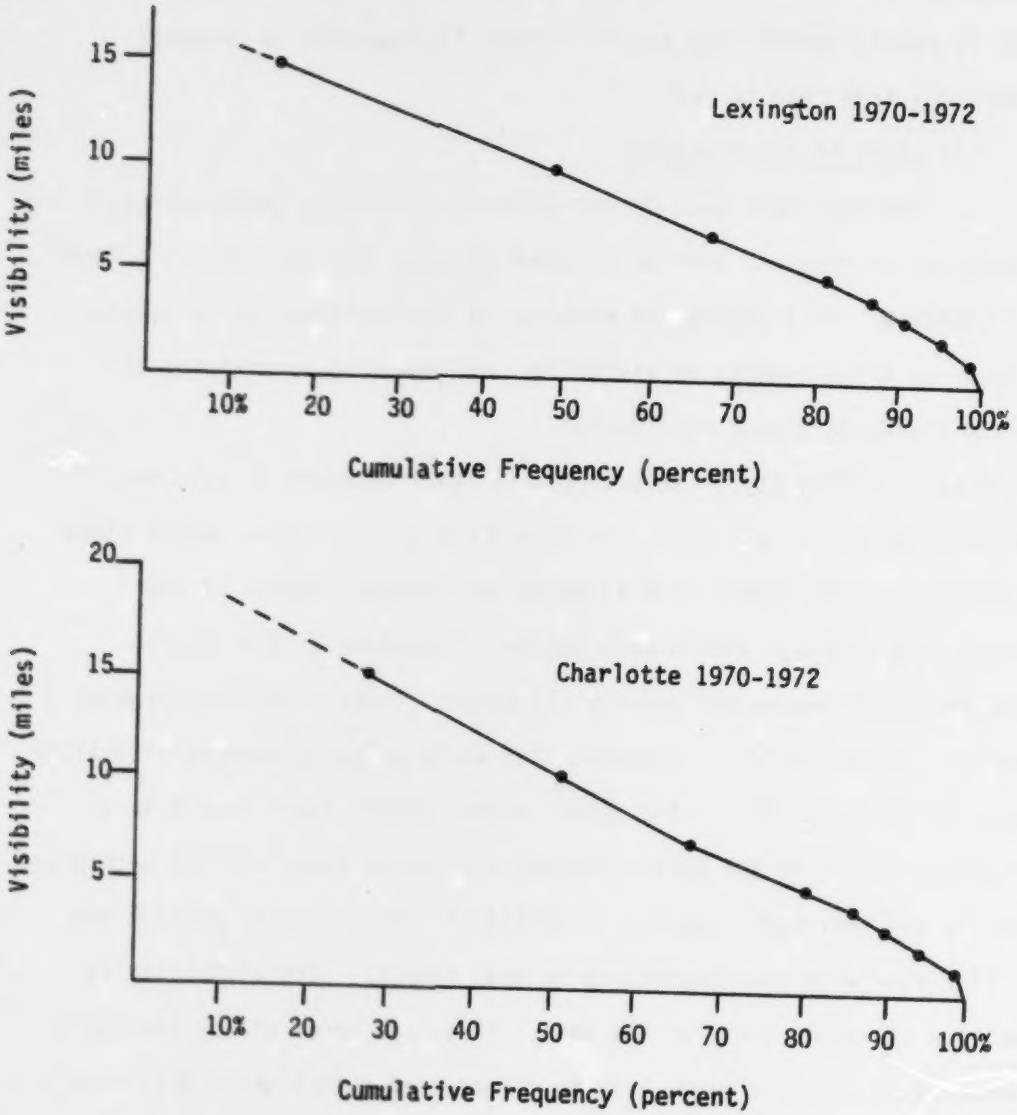

Figure 7-3. Cumulative frequency distributions of visibility at urban/suburban locations (Trijonis *et al.*, 1978b).

standard should be expressed in statistical form such that seasonal (quarterly, e.g., summer) values are compared for multiple years. This tends to smooth variability due to natural fluctuations in seasonal meteorology from year to year.

iii) Level of the Standard

The available data on the value of visibility (both aesthetic and transportation related) provide no clear guidance for an acceptable level of a standard. In arraying the elements of the decision, it is helpful to focus on those aspects of visibility impairment where national air quality standards appear most useful.

A major difficulty in selecting a national standard is evidence that both the extent of visibility impairment and the value people place on visibility vary widely with affected populations, region of the country, and settings within each region. Therefore an air quality standard cannot reasonably address all facets of the visibility problem equally. In particular, a standard that would maintain current visibility levels in the rural West ($< 3\text{-}5\ \mu g/m^3$) might require lower than natural background levels in the East. Fortunately, other Clean Air Act mechanisms directly and indirectly address visibility.* These clearly provide the Administrator with the flexibility to deal directly with visibility in sensitive non-urban areas of the West. Thus, the level of any secondary fine particle standard might best be directed at establishing desirable visibility goals for those regions in the East affected by large scale regional haze of multistate origin and those major western urban centers affected by haze predominately of local origin.

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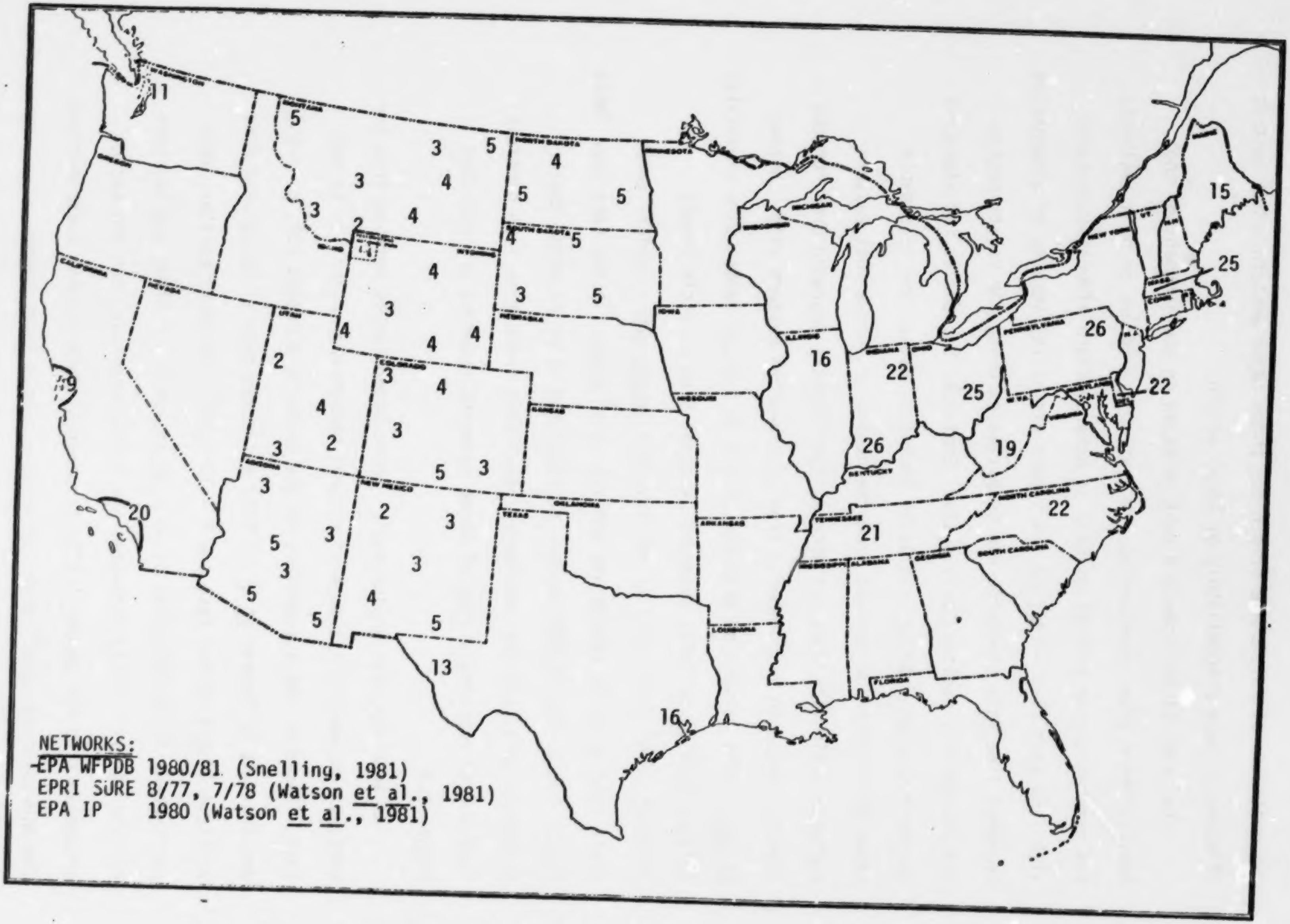
*The prevention of significant deterioration (PSD) provision of the CAA establishes limits on particle (and other pollutant) increases over "baseline" conditions. Certain "class I" areas (e.g., national parks, wilderness areas) where visibility is an important value receive special PSD and visibility protection. The interaction among these and other CAA regulatory mechanisms that affect visibility is further discussed in Chapter 7 of Protecting Visibility: An EPA Report to Congress (EPA, 1979).

The level of any standard must be based on the judgment of the Administrator after considering the available evidence on both aesthetic and transportation effects as well as other welfare effects associated with fine particles. Long or short-term visibility impacts of alternative seasonal fine mass standards can be estimated using the relationships and assumptions outlined previously. Empirical boundaries for standards can be estimated from the available technical data. One reasonable upper bound might be derived from the desire to ensure that eastern regional visibility does not become significantly degraded over current levels. Based on measurements from 12 non-urban eastern sites (Watson et al., 1981) as well as estimates based on typical summer time visibility (Figure 7-1b), spatially averaged seasonal fine particle levels of 22-25 $\mu g/m^3$ would tend to maintain the status quo. Typical urban scale fine particle levels are about 5 $\mu g/m^3$ higher in the East (see Table 4-3). Due to the limited number of sites and years of data, these estimates obviously are uncertain. Available summertime fine particle levels from non-urban sites of three research networks are depicted in Figure 7-4.

An absolute, but unattainable, lower bound can be derived from the criteria document's estimate of natural background visibility in the East. Based on the assumptions in the criteria document (CD, p. 9-63) and the data of Ferman et al. (1981) estimated natural background fine particle levels in the East are 6 - 10 $\mu g/m^3$. As noted earlier, this corresponds to visibilities of 60 - 80 km, or 3 - 5 times greater than current levels. These estimates are quite uncertain, but the basic assumptions do not appear likely to underestimate natural contributions. The mid-point (8 $\mu g/m^3$) might serve as a single lower bound.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NETWORKS:
EPA WFPDB 1980/81 (Snelling, 1981)
EPRI SURE 8/77, 7/78 (Watson et al., 1981)
EPA IP 1980 (Watson et al., 1981)

Figure 7-4. Summertime Fine Particle Levels - Non-Urban Sites

The upper and lower bounds for a seasonal (three month) fine particle standard thus derived are 8 - 25 $\mu g/m^3$, spatially averaged over distances on the order of 16 - 50 km. The range is perhaps surprisingly narrow. Nevertheless, as illustrated in Figure 7-2, visibility is extremely sensitive to fine particle levels in this range. If visibility benefits were proportional to "typical" annual visual range, then marginal benefits would increase with decreases in fine particle concentrations.

B. Materials Damage and Soiling

1. Description of Effects

The deposition of airborne particles can become a nuisance, degrade aesthetics and material usage through soiling and may contribute directly, or in conjunction with other pollutants, to structural damage by means of corrosion or erosion. The nature of these effects is discussed below and the relative importance of particle size, composition, and other environmental factors such as moisture, temperature, sunlight, and wind also is considered.

a) Materials Damage

Particles affect structural materials principally by promoting and accelerating the corrosion of metals, the degradation of paints, and the deterioration of building materials such as concrete and limestone. Particles exhibit these effects because of their electrolytic, hygroscopic, and/or acidic properties, and their ability to sorb corrosive gases. Our review suggests that only chemically active fine mode or hygroscopic coarse mode (mainly sea or road salt) particles materially contribute to such effects (CD, p. 10-41).

While particles have been qualitatively associated with damage to paints and building materials, there are insufficient data at present to relate such effects to specific pollutant levels. With respect to the corrosion of metals, field studies (Mansfield, 1980; Haynie and Upham, 1974; and Upham, 1967) have not established a quantitative relationship between particles and corrosion (CD, p. 10-41). This may be explained by the presence of sulfur dioxide which, in combination with sufficient moisture, dominates the corrosion rate to such extent that an independent effect of particles is not evident (Yocom and Grappone, 1976). The available data do not clearly suggest major effects of particles on materials for concentrations at or below the ranges recommended for the primary health standards, but to the extent damages occur, they are more likely associated with active fine particles. Potential materials damage effects thus may provide ancillary support for a fine particle secondary standard.

b) Soiling

Soiling is the accumulation of particles on the surface of an exposed material resulting in the degradation of its appearance. When such accumulations produce sufficient changes in reflection from opaque surfaces and/or reduce light transmission through transparent materials, the surface will become perceptibly dirtier to the human observer. At higher loadings, the effect will be detectable by touch. In a subjective perception/values study, Hancock *et al*. (1976) determined that "dirtiness" for a flat surface would occur when the accumulation of dust particles is sufficient to cover 0.7% of the material's surface under conditions of maximum contrasts (e.g., black on white or vice versa). Similar studies have not been reported for other soiling effects.

Despite the numerous observations that airborne particles soil a wide range of man-made materials, there is only limited information available with respect to the size and composition of the culpable particles. In general, the soiling of fabrics and vertical surfaces has been ascribed to fine particles, particularly dark carbonaceous materials. Soiling of horizontal surfaces may result from deposition of a wide range of particles, including coarse mode dusts.

Theoretically, coverage of horizontal surfaces will be related to particle surface area and deposition velocity. Particle surface area per unit mass decreases linearly with diameter (assuming spherical particles), while, under quiescent conditions, deposition velocity increases with the square of the diameter. Under such conditions, large particles would result in more soiling than an equivalent mass of smaller particles. Although second order effects may enhance fine particle deposition relative to larger particles, the deposition velocity data in Chapter 6 of the criteria document still suggest substantially higher deposition on horizontal surfaces for particles larger than 10 μm than for smaller particles.

The increasing soiling potential associated with increased particle size is mitigated by lighter particle color, smaller transport distance from sources, and markedly lower penetration of larger particles to indoor surfaces (relative to smaller particles). Alzona _et_ _al_. (1979) concluded, on the basis of their study of indoor/outdoor element ratios, that indoor exposure to outdoor dust would be reduced by two-thirds with the doors and windows partially closed (CD, p. 5-127). Because these conflicting factors have not been quantitatively evaluated, it is not possible to make clear particle size divisions with respect to soiling of horizontal surfaces.

The time interval that it takes to transform horizontal and vertical surfaces from a clean to a perceptibly dirty state is generally determined by particle composition and the rate of deposition. This process also is influenced by the location and spatial alignment of the material, the texture and color of the surface relative to the particles, and meteorological variables such as moisture, temperature, and wind speed.

2. Quantitative Associations

a) Soiling/Property Values

The effect of particles on aesthetic quality depends in part on human perception of pollution. The reduction of aesthetic quality may arise from the soiling of buildings or other objects of historical or social interest or by the mere dirty appearance of the neighborhood. A number of studies have indicated that such perceptions of neighborhood degradation are revealed indirectly through effects on the value of residential property. That is, when residential properties similar in other respects are compared, the properties in the more highly polluted areas typically have lower value.

Freeman (1979a), reporting on 14 property value studies that used particulate matter or dustfall as one of their pollutant measures, noted that the results generally supported the premise that property values are affected by the full range of particle pollution. He cautioned, however, that direct comparison of the monetary results is not possible since the studies cover a number of cities, use different data bases, empirical techniques, and model specifications.

The extent to which the city specific results represent soiling as opposed to perceptions of the effects of particles on health and visibility is not clear. Therefore, the results of studies such as those described above cannot provide reliable quantitative estimates of the effects of soiling on property values.

b) Soiling/Household Cleaning

Airborne particles soil a wide range of materials in all sectors of the economy. Assuming that these sectors are not as well off in a dirtier state as a cleaner one, soiling will result in an economic cost to society. While the household sector has been examined by a number of investigators, their results have been questioned because of methodological problems and their failure to appropriately address particle size, composition, and deposition rates. As a result, no single study has produced a completely satisfactory estimate of soiling costs for the household sector. It is unfortunate that little or no effort has been expended to account for soiling costs in the commercial, manufacturing, or public sectors. For example, preliminary results from Manuel et al. (1981) suggest that the omission of the manufacturing sector alone could result in a significant understatement of soiling costs due to particle pollution.

In its review of effects of household soiling, the criteria document relied principally on Booz, Allen, Hamilton (1970), Watson and Jaksch (1978, 1982), and Freeman (1979b) to derive estimates for both household soiling costs in 1970 and the benefits that accrued in 1978 as the result of reduced total suspended particulate (TSP) levels. For the year 1970, the estimate for amenity loss due to exterior household soiling was estimated to range from 1 to 3.5 billion dollars (1978 dollars). The 14 $\mu g/m^3$ reduction in U.S. annual TSP levels between 1970 and 1978 was estimated to have resulted in an annual benefit for the year 1978 of 0.2 to 0.7 billion dollars or 14 to 50 million dollars for each $\mu g/m^3$ of reduction (CD, p. 10-73).

These estimates obviously are rough and mainly provide an indication that substantial costs may be associated with particle related soiling. Some of the important limitations include:

1) The exclusion of interior household soiling costs from these estimates would tend to bias the results downward since the analysis did not address the soiling effects of those smaller particles that readily penetrate the indoor environment (CD, p. 5-131).

2) Even for estimating exterior soiling costs, the use of TSP may be a poor pollutant indicator. As noted earlier, the soiling of vertical surfaces generally have been ascribed to fine particles while the soiling of horizontal surfaces may result from the deposition of a wide range of particles, including coarse dusts. Moreover, national average TSP readings may be a poor surrogate for actual exposures.

3) The basic data on costs and frequency of performing household cleaning and maintenance tasks used in the studies relied on were derived for Philadelphia and may not be readily transferable to other parts of the country.

4) The studies relied on had both positive and negative biases which cannot be assumed to cancel out, thus adding additional uncertainty to the final estimates.

3. Staff Recommendations

It is clear that at high enough concentrations, the full size range of particles including dustfall can contribute to soiling, become a nuisance and result in increased cost and decreased enjoyment of the environment.

The available data are limited, however, and do not permit any definitive findings with respect to the economic costs or other response functions associated with particle soiling or provide clear quantitative relationships between ambient loadings and soiling. The limited data do suggest, however, that soiling costs can be significant.

The key question is whether the recommended range of primary standards would provide adequate protection against these effects. Much of the range of primary standards would appear to permit increases in TSP loadings. Thus, the staff recommends consideration of the economic and other effects associated with soiling and nuisance when determining whether a secondary standard for TP or for TSP or other large particle indicator is desirable to supplement whatever soiling benefits accrue from the primary health standards. Because fine particles are a significant contributor, soiling effects also should be considered in determining whether a fine particle secondary standard is needed. Given the uncertainties in the data, recommendations on ranges of interest are not appropriate at this time.

C. Vegetation Damage

1. Description of Effects/Relationships

Controlled laboratory and crop field studies indicate that current levels of "general" particulate matter are relatively unimportant from the standpoint of vegetation damage. Controlled laboratory studies have associated vegetation damage only with very high particle dustfall rates of insoluble particles (Lerman and Darley, 1975; NAS, 1977a). Field studies near emission sources have reported injury to vegetation following exposures to high dustfall rates of general particulate matter or specific phytotoxic substances that may be found in particulate matter (EPA, 1975a; Lerman and Darley, 1975; NAS, 1977a).

Controlled exposure studies of insoluble dusts (e.g., cement dust) indicate that the accumulation of particulate matter on leaf surfaces can result in damage to vegetation when particle dustfall rates are very high. Although some leaf injury from insoluble particles may result from reduced light transmission to the leaf surface following heavy particle accumulation on leaf surfaces, the more likely cause of leaf injury from insoluble particles is from particle lodging/penetration in stomatal apertures. Partial stomatal occlusion by particles could alter normal gas exchange patterns resulting in reduced carbon dioxide uptake during the day when plants fix carbon dioxide into carbohydrates. Since most stomatal apertures are less than 10 μm long and 4 μm wide when open (Esau, 1960), particle lodging/penetration should become more efficient as particle sizes approach those that can fit into stomatal apertures. Plant exposure to 0.6 g/m^2/day (~500 $tons/mi^2$/mo) of cement dust ("most" of the dust represented particles less than 10 μm) for 8-10 hours/day is the lowest exposure regime reported to produce a physiologic response (a reduction in carbon dioxide uptake). Foliar injury was reported at higher exposure regimes in this study (Darley, 1966).

Field studies of the effects of cement dust on citrus trees have reported foliar injury, damage to fruit, and reduced yield at high dustfall rates (about 290 $tons/mi^2$/mo) that were observed around 1900 (Parish, 1910). Since deposition rates substantially less than about 75 $tons/mi^2$/mo have generally been observed even in the more polluted urban areas of the United States in the 1960's (DHEW, 1969), it is unlikely that plants would be injured by the lower particle deposition likely to be found today. Exceptions could occur in proximity to strong point sources.

Plants have, however, been shown to respond to exposure from relatively high concentrations of specific phytotoxic substances that are sometimes found in particulate matter. The entrance of such particles into plant tissues is facilitated when such particles are small enough to penetrate stomatal apertures or when soluble materials are deposited in the soil and absorbed by plant roots (Brady, 1974). In addition, some components of particulate matter (especially soluble fractions) that are deposited on leaf surfaces could subsequently penetrate to the leaf interior. Substance specific toxicity has been reported in plants following exposure to arsenic (Linzon, 1977), boron (Kupra and Kohut, 1976), fluorides (Lerman and Darley, 1975), soot (Lerman and Darley, 1975), zinc (Yopp *et al*., 1974), and other substances.

2. Staff Recommendations

Recognizing that specific toxic particulate substances that may affect public welfare generally are found only in trace quantities except near specific sources, regulatory mechanisms other than the national ambient air quality standards (e.g., section 111(d)) are used to regulate specific toxic substances. The effects of fine acid aerosols on vegetation should be considered as a part of the acidic deposition phenomenon.

In view of the lack of any clear effect of particle exposure that is not composition specific on plants at current particle levels, the staff concludes that a secondary particle standard is not needed to protect vegetation.

D. Personal Comfort and Well-Being

Because the effects of thoracic deposition, including the production of health related symptoms causing discomfort, are incorporated in the primary standard discussion and recommendations, this assessment will focus on direct effects of extrathoracic and extra-respiratory particle deposition on personal comfort.

1. Description of Effects, Quantitative Data

A controlled exposure study found that discomfort is proportional to the concentration of dust and that symptoms can be produced by "inert" (carbon impregnated plastic) particles deposited in the nose and throat (Andersen *et al.*, 1979). Subjects were young, healthy adults characterized as nose breathers, and were at rest during exposure. Symptoms from extrathoracic particle deposition included dryness of the nose, pharynx, and throat and other mild symptomatic responses. Particle size was about 9-11 μm (MMAD) with 30% of the mass larger than 12.4 μm. The lowest reported concentration associated with symptomatic effects was 2 mg/m^3 (Andersen *et al.*, 1979). Although the symptomatic response to dust exposure was noted one to two hours following the beginning of the dust exposure, available subjective information indicates that discomfort was mild.

In high enough concentrations, large particles such as wind blown dust or cinders also might cause discomfort through impaction on the eye or skin (extra-respiratory deposition). The scientific literature summarized in the criteria document, however, provides no basis for evaluating the concentrations at which such effects might occur.

2. Staff Recommendations

The available information suggests that particles might affect personal comfort at total particle concentrations in excess of 2 mg/m^3 for short exposure (2 hour) periods. The data are, however, extremely limited, and not unequivocally associated with larger particles. Although it is reasonable to suggest that high dust concentrations are a nuisance that affects personal comfort, the available data provide no clear evidence as to what concentrations of larger particles not included in the primary standards might be of concern. The data in the criteria document in this area thus provide little support for a TSP or other large particle indicator secondary standard based on soiling and nuisance effects discussed in Section VII.B.

E. Acidic Deposition

On August 20-21, 1980, the Clean Air Act Scientific Advisory Committee (CASAC) concluded that the issue of acidic deposition was so complex and important that a significantly expanded and separate document would be necessary if NAAQS were to be selected as a regulatory mechanism for control of acidic deposition. CASAC noted that a fundamental problem of addressing acid deposition in a criteria document is that acidic deposition is produced by several pollutants (including oxides of nitrogen, oxides of sulfur, and acidic fine particles). Consequently, a document on acidic deposition would include various pollutants contributing to wet and dry deposition. The Committee also recommended that a revised version of the acidic deposition chapter be retained in PM/SO_x and NO_x criteria documents. In response to these recommendations, EPA is in the process of developing an acidic deposition document that will provide a more comprehensive treatment of this subject. Thus, the issue will not be directly addressed in this staff paper.

F. Summary of Staff Conclusions and Recommendations

Major staff conclusions and recommendations made in Section VII.A-E are summarized below:

1a) Impairment of visibility by fine particles over urban to multi-state regions clearly affects public welfare. Fine particles or major constituents thereof also are implicated in climatic effects, materials damage, soiling, and acid deposition. Neither the current TSP secondary standard nor the recommended ranges of TP standards will protect visibility in an effective manner. The staff, therefore, recommends consideration of a fine particle secondary standard, based primarily on the relatively well defined quantitative relationships between fine mass and visibility.

b) If a fine particle standard is selected, a seasonal (calendar quarter) averaging time could provide a statistically stable target and yet achieve most short or long-term visibility goals. Consideration should be given to specifying a spatial average of three or more monitors placed at distances on the order of 16-50 km.

c) Despite the fact that the public is concerned about visibility and is willing to pay something for clean air, quantitative bases for evaluating visibility goals have not been established. Therefore, the level of any standard must be based on the judgment of the Administrator after consideration of aesthetics, transportation, as well as non-visibility related effects. The staff recommends that any national standards focus on welfare effects associated with multistate eastern regional (and western urban) haze. Such standards would not of themselves protect sensitive scenic areas of the West, but these areas are directly and indirectly addressed by other provisions of the Clean Air Act.

d) Empirical ranges for standards can be derived from approximate estimates of eastern natural background and current summertime fine particle levels. The range thus derived is 8 - 25 $\mu g/m^3$, seasonal and spatial average. The upper portion of the range would tend to maintain the status quo in the East. Because the lower portion of the range approaches natural background levels, standards set at the lower levels would be, in all practicality, unattainable in most of the Eastern U.S.

e) Because regional fine particles in the East appear to be influenced most strongly by sulfates, adoption of a fine particle standard would trigger a substantial departure from current approaches to particle control strategies. The evidence suggests that multistate control of regional sulfur oxide emissions might be needed to reduce fine particle levels. Thus, fine particle/visibility-climate effects are linked to acid deposition, and these problems would likely be ameliorated by similar control strategies. Addressing these welfare effects with a common standard or control strategy is likely to be more efficient than establishing separate control approaches for each. Appropriate scientifically based targets and control strategies for acid deposition are not yet available.

2) Although potential effects on climate support the consideration of a fine particle standard, quantitative relationships are not well enough developed to provide the principal basis for selecting the level of the standard.

3) Consideration should be given to soiling and nuisance effects in determining whether a secondary standard for TP or for TSP or some other large particle indicator is desirable to supplement the primary health and secondary fine particle standards. The available data base on such effects is, however, largely qualitative. Therefore, the basis for selecting a particular level for a secondary TP or TSP standard is a matter of judgment.

4) While chemically active fine mode and hygroscopic coarse mode particles have been qualitatively associated with materials damage, the available data do not clearly suggest major effects of particles on materials for concentrations at or below the ranges recommended for the primary health and secondary visibility standards. Therefore, a secondary standard based solely on materials damage is not recommended.

5) The staff concludes that a secondary particle standard is not needed to protect vegetation.

6) The acid deposition issue will not be addressed directly in the review of the particulate matter standards.

APPENDIX A. FACTORS THAT INFLUENCE DEPOSITION AND CLEARANCE OF PARTICLES

This material briefly summarizes subject related and environmental factors that affect deposition and clearance of particles, and supports the discussion in Section V A.1 of this paper.

A. Inhalation Patterns

The most important aspects of inhalation pattern are flow rate, breathing frequency, and whether breathing is through nose, mouth, or both. Nasal breathing results in nearly complete extrathoracic deposition for all particles larger than about 10 μm, with removal efficiency decreasing to less than 10% for particles less than 1 μm (CD, p. 11-23). In contrast, extrathoracic deposition of 10 μm and 1 μm particles for mouth breathing is about 65% and 2%, respectively (CD, p. 11-23). As illustrated in Figure 5-1, mouth breathing enhances the availability of > 1 μm particles for deposition in the alveolar and tracheobronchial regions. Because the mouth can release substantial quantities of endogenous ammonia, mouth breathing may provide partial protection from airborne acids by ammonia neutralization (Larson et al., 1977).

Except in the case of blockage of the nasal passages by mucus or other obstruction, typical "mouth" breathing is better characterized as "oronasal" breathing, since the larger portion of the inspired air does pass through the nasal passages. Therefore, experimental deposition studies as summarized in Figure 5-1 that use mouth piece only breathing may tend to overestimate thoracic deposition for oronasal breathing at rest. Approximately 15% of the population are habitual oronasal or mouth only breathers (Saibene et al., 1978; Niinimaa et al., 1981). Anyone may shift to oronasal breathing during conversation, singing, illness related congestion, and exercise. Most people shift to oronasal breathing at flow rates greater than 30-35 l/minute (Ninnimaa et al., 1981), which is equivalent to moderate exercise as in walking briskly or bicycling (Cotes, 1979). At these flows, the quantity of air passing through the mouth is greater than that used in resting, mouth only deposition studies. Thus, results of such studies might not overestimate deposition for exercising conditions with oronasal breathing.

Increased flow rates also increases impaction related deposition in all regions and apparently speeds mucuciliary clearance (Wolff et al., 1975). The generally increased extrathoracic and tracheobronchial deposition efficiency in the 2-10 μm range, reduces the alveolar deposition efficiency in this same range (Heyder et al., 1980; Stahlhofen et al., 1980). Nevertheless, because of increased intake, as well as a shift to mouth breathing, total dose to the alveolar region may increase with exertion related increases in flow rate. For example, Heyder et al. (1980) found that the alveolar deposition efficiency (mouth breathing) for a 5 μm particle was about 0.4 at a flow rate of 250 cm^3. At three times the flow rate, with constant breathing frequency, the efficiency dropped to 0.3, but alveolar dose/unit time would be increased by a factor of about 3 x 0.3/0.4 = 2.2.

B. Subject Airway Dimensions, Disease State

Size, shape and branching of the airways and other airspaces markedly affect deposition of particles (Palmes and Lippmann, 1977). Evaluation of available data reproducibility of individual deposition curves (Chan and Lippmann, 1980) and studies of identical twins (Bohning et al., 1975) suggest that much of the variability observed in various deposition studies is largely due to anatomical differences among subjects. Among normal adults total respiratory deposition can vary by a factor of 4 (lowest to highest) (Giacomelli-Matoni et al., 1972). Although the criteria document reports no deposition studies on children, it is reasonable to expect that the smaller airway dimensions in children would result in relatively greater impaction related deposition in the tracheobronchial and extrathoracic regions than in normal adults.

Disease states or other conditions that constrict, inflame, or cause mucous build-up in the airways can substantially increase tracheobronchial deposition (Lippmann et al., 1971). Otherwise normal cigarette smokers have somewhat increased bronchial deposition (Lippmann et al., 1971), and greatly enhanced bronchial deposition was seen in chronic bronchitics and asymptomatic asthmatics (Albert et al., 1973). Over a size range of 1.5 to 6 μm, deposition was up to a factor of 5 greater than the upper bound of the normal range.

Clearance rates for deposited materials also varies substantially among normal healthy adults (Yeates et al., 1975). Studies of bronchial clearance in patients with chronic lung disease have reported transport rates which were faster, slower, and similar to those of healthy adults (NAS, 1977a). Within the tracheobronchial region the nature of the branching airways also results in nonhomogeneous deposition patterns. Of particular significance are the observations and calculations indicating increased deposition at "hot spots" located around the carina of airway bifurcations (Bell and Friedlander, 1973; Schlesinger and Lippmann, 1978). The latter authors were able to demonstrate a correlation of upper airway deposition patterns and reported incidence of bronchogenic carcinoma. Effects at the bifurcations also may be enhanced due to potentially slower clearance rates (Hilding, 1957).

C. Aerosol Composition

Aerodynamic diameter, chemical composition, and shape are among the more important particle specific parameters affecting deposition. The importance of initial aerodynamic diameter is illustrated in Figure 5-1. A substantial fraction of ambient aerosol consists of hygroscopic acids or salts (e.g., sulfuric acid and sulfates, sea salt). As noted in Section IV, as humidity increases, these substances accumulate water and increase in size. The respiratory tract humidity of inspired air nears 100% in the vicinity of the nasopharyngeal cavity, and the residence time of smaller aerosols is sufficient to allow growth by a factor of 2 to 4 (Ferron, 1977; Tang, 1980). Evidence of such growth is provided by Utell et al. (1981) who measured total respiratory tract deposition after oral inhalation of several sulfates and sodium chloride, initially of 0.5-1.0 μm aerodynamic diameter. Total deposition efficiency (.55 - .65) was much higher than expected for the initial size, and was in the range expected for 2-4 μm particles (Figure 11-4, CD).

Growth of ambient hygroscopic aerosols in the fine mode would be expected to enhance deposition in all three regions of the respiratory tract. Because total deposition increases rapidly with sizes above about 1-2 μm, increased deposition resulting from hygroscopic growth of a factor of 2 to 4 would be most important for accumulation mode particles larger than about 0.4 μm. Neutralization of acid aerosols by endogenous ammonia decreases with larger particle size (Larson et al., 1982).

Gaseous components of pollutant atmospheres, as well as irritant particles can affect particle deposition by causing bronchoconstriction (Lippmann, 1977). As noted above, airway constriction tends to markedly increase tracheobronchial deposition. Among the more prevalent irritant gases that occur in combination with particles in U.S. atmospheres are ozone, sulfur dioxide, nitrogen dioxide, and formaldehyde. As discussed in Section V.A., a number of particulate substances may cause bronchoconstriction.

APPENDIX B. EVALUATION OF EVIDENCE FOR EFFECTS OF CONCERN

This section discusses and evaluates the key studies providing evidence on the kinds of effects that might reasonably be anticipated to result from exposure to atmospheric particles. As outlined in Section V-B, the principal effects of concern include:

1) Respiratory mechanics and symptoms;
2) Aggravation of existing respiratory and cardiovascular disease;
3) Clearance and other host defense mechanisms;
4) Morphological alterations;
5) Carcinogenesis; and
6) Mortality.

Because of the complexity of the data base and lack of clear quantitative relevance to general populations, the criteria document lists but does not discuss a number of occupational studies and reviews that may be of qualitative interest. The staff has evaluated a number of these documents and has selected those of interest for discussion in this section. These results are illustrative of the kinds of responses that can be elicited in groups usually characterized as normal, adult healthy males after prolonged high exposures to a variety of different "inert" dusts and reactive particle classes. Although significant weight is not given to these studies in the following discussions, they are listed in Table B-1 as supportive material.

A. Respiratory Mechanics and Symptoms

A number of functional measurements are used to indicate altered mechanical and flow attributes of the respiratory tract following pollutant exposures. Effects on respiratory mechanics and function can range from mild transient changes of little direct health significance to incapacitating impairment of breathing. Mild effects in normal subjects may indicate potentially more

TABLE B-1. RESPIRATORY DISEASES AND RELATED IMPAIRMENTS ASSOCIATED WITH OCCUPATIONAL EXPOSURE TO PARTICLES

Biological Endpoint	Particle	Reference
1. Deterioration of lung capacity/ Pulmonary Function Decrement	Coal Dust	Lapp et al., 1972 Morgan, 1978
	Carbon Black	Valic et al., 1975
	Cotton, Flax and Hemp Dust	NIOSH, 1975b
	Grain Dust	Dosman et al., 1980
2. Bronchitis	Coal Dust	Morgan, 1978
	Grain Dust	Dosman et al., 1980
3. Asthma	Grain Dust	Dosman et al., 1980
4. Pneumoconiosis	Coal Dust	Morgan, 1978
	Slate Dust	Glover et al., 1980
4a. Silicosis	Free Silica Dust (sandblasting)	Ziskind et al., 1976
	Kaolin	Ziskind et al., 1976
	Mica and Quartz (slate)	Ziskind et al., 1976
4b. Silicosis and Mycobacterial Infection (e.g., Tuberculosis)	Free Silica Dust	Ziskind et al., 1976
5. Pulmonary Fibrosis	Granite Dust	Craighead and Vallyathan, 1980
	Asbestos	NIOSH, 1976
6. Byssinosis	Cotton, Flax and Hemp Dust	NIOSH, 1975b
7. Mesothelioma and Respiratory Cancer	Asbestos	NIOSH, 1977
	Arsenic	NIOSH, 1975a
	Coke Oven Emissions	NAS, 1972

serious responses in more sensitive subjects. Symptomatic effects such as coughing, wheezing, or bronchospasm also vary in severity, but at minimum are indicative of a biological response. This section discusses the effects of specific particulate components and community aerosols on respiratory mechanics and symptoms.

1. Sulfates and Other Water Soluble Aerosols

Sulfate compounds are among the most widely tested particulate components in animal and human controlled exposure studies. Studies of these and other soluble aerosol substances provide some clues to their relative irritant potential and factors that affect responses.

Unfortunately, animal studies of sulfur-containing acid aerosols (sulfuric acid, ammonium bisulfate) may be seriously compromised by the presence of high chamber ammonia which may partially or fully neutralize the acidity. The problem of neutralization is most serious in long-term studies where both exhalation (Larson et al., 1977) and the accumulation of excrement (Malanchuk et al., 1980) contribute to chamber ammonia. Short or long term studies that do not explicitly deal with this problem by measurement or other approaches do not provide reliable quantitative assessments of the effects of acid aerosols alone. In some short term studies, neutralization may be reduced to some extent by systems that use head only exposures and should not be compromised by excrement ammonia (e.g., Amdur and Mead, 1955). Even in well designed studies, however, it is difficult to determine the reliability of reported sulfuric acid and bisulfate levels, due to possible differences in exhaled ammonia between animals and people (Larson et al., 1977).

A number of experiments have been conducted in guinea pigs to rank the relative respiratory irritancy potential (as measured by airway resistance) of sulfuric acid and other sulfates by size and species. Amdur and co-workers report that the relative irritancy increases as particle size decreases but only at lower concentrations (Amdur, 1958). However, observations of animal response at higher concentrations than in this study indicate that increases in airway resistance are greater for 2.5 than for 0.8 μm diameter particles. Other investigators comparing the relative irritancy of 2.7 and 0.8 μm sulfuric acid aerosol report that death rates are higher for larger particles (Pattle et al., 1956).

Amdur and co-workers (1978a) report that the irritancy potential of various sulfates roughly follows their acidity, with the exception that NH_4HSO_4 was less irritant than $(NH_4)_2SO_4$. Clear distinctions between these substances are difficult to resolve because of differences in particle size and concentrations and the potential neutralization problem. Although Amdur and co-workers (1978b) report changes in pulmonary mechanics following a single hour of exposure as low as 100 μg/m^3 H_2SO_4, other investigators have not been able to produce similar changes in guinea pigs, dogs, sheep, and donkeys below at least 1,200 μg/m^3 (Sackner et al., 1978; Schlesinger et al., 1978; Silbaugh et al., 1980). Differences in animal susceptibility (CD, p. 12-79) and experimental techniques may be responsible for these observed differences. Within Amdur's studies, some guinea pigs were apparently more susceptible than others. One interesting feature of the Amdur work is the apparently linear dose response curve for some sulfate species (Figure B-1). No evidence exists for a "threshold" of response in this animal model.

A number of recent studies have examined human respiratory mechanical responses to sulfuric acid and other sulfates (Table B-2). In essence, no

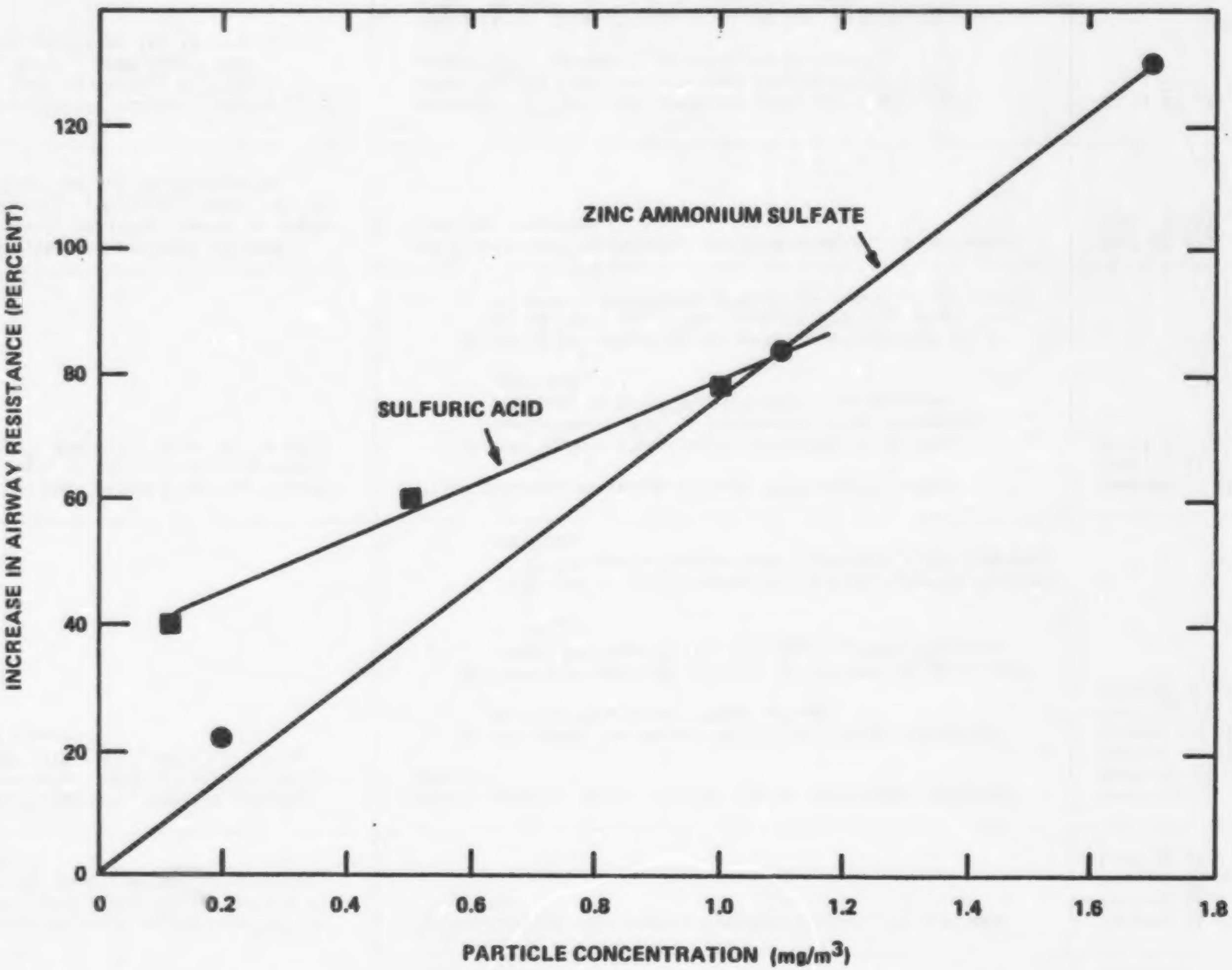

Figure B-1. Concentration response relationship for two sulfate areosols in guinea pigs, one hour exposures (Amdur and Corn, 1963; Amdur et al., 1978b). Zinc ammonium sulfate is included for illustrative purposes. It is not thought to be a commonly found air pollutant.

TABLE B-2. EFFECTS OF SULFURIC ACID AND OTHER SULFATES ON RESPIRATORY MECHANICS

Exposure	Observations	Reference
Healthy adult subjects exposed various routes*, activity levels to sulfuric acid aerosol 10-300 μg/m^3, MMD 0.1-1.5 μm for 10-240 minutes	No significant respiratory function changes, no symptoms reported	Sackner *et al.*, 1978 Avol *et al.*, 1979 Leikauf *et al.*, 1981 Kerr *et al.*, 1981
Healthy adult subjects, exposed various routes*, activity levels to sulfuric acid aerosol, 350-1000 μg/m^3, MMD 0.1-1.5 μm for 10-120 minutes	Mixed results; three studies report respiratory function changes: a) 350 μg/m^3--increased respiratory rates, decreased maximum flow rates, tidal volumes b) Increased symptoms (throat irritation, dryness, and cough) between 220 and 940 μg/m^3, slight decrease in FEV$_{1.0}$ c) 1000 μg/m^3-- small reduction in flow enhanced response to bronchoconstrictive agent (carbachol). No symptoms reported	Amdur *et al.*, 1952 Newhouse *et al.*, 1978 Sackner *et al.*, 1978 Leikauf *et al.*, 1981 Utell *et al.*, 1981 Horvath *et al.*, 1981
Asthmatic subjects exposed various routes*, activity levels to sulfuric acid aerosol 10-1000 μg/m^3, MMD 0.1-1.5 μm for 10-120 minutes	Mixed results; one study reports functional changes: a) 1000 μg/m^3--significant reduction in air way conductance, FEV$_{1.0}$, expiratory flow; enhanced response to bronchoconstrictor. No symptoms reported. b) 100 μg/m^3--enhanced response to bronchoconstrictor in two most sensitive subjects (Utell); suggestion of airway resistance effects in 2 asthmatics (Avol).	Sackner *et al.*, 1978 Avol *et al.*, 1979 Utell *et al.*, 1981
Normal and asthmatic subjects exposed various routes*, activity levels to several sulfates NH_4HSO_4, $(NH_4)_2SO_4$, $NaHSO_4$ 85-100 μg/m^3 MMD 0.4-1 μm for 16-150 minutes	No significant respiratory function changes, no increased symptoms reported	Avol *et al.*, 1979 Utell *et al.*, 1981
Normal and asthmatic subjects exposed orally, at rest to (H_2SO_4, NH_4HSO_4, $(NH_4)_2SO_4$, $NaHSO_4$) 1000 μg/m^3, MMD 0.5-1 μm dry particles for 16 minutes	Normals: All sulfates produced small but significant reduction in flows and enhanced response to bronchoconstrictor, generally in relation to acidity. Asthmatics: Same effects as noted for normals, in addition H_2SO_4, NH_4HSO_4 resulted in decreased conductance, FEV$_{1.0}$.	Utell *et al.*, 1981

*e.g. Nose only, mouth only, free breathing.

direct changes in respiratory function have been reported for short term exposure to normals and asthmatics at levels below 350 $\mu g/m^3$, and only one study has reported symptomatic responses in these groups at levels below 1000 $\mu g/m^3$.

Both normals and asthmatics exposed to 1000 $\mu g/m^3$ of sulfuric acid, ammonium bisulfate, ammonium sulfate, and sodium bisulfate responded with small but significant reductions in maximum expiratory flow volume at 60% total lung capacity (TLC) and partial expiratory flow-volume at 40% and 60% TLC (Utell et al., 1981). At this concentration, sulfuric acid and ammonium bisulfate also produced significant reductions in airway conductance and forced expiratory volume at 1 second. All sulfates also enhanced the response of normals and asthmatics to subsequent bronchoconstrictor challenge. In the two asthmatics most sensitive to sulfate at 1000 $\mu g/m^3$, sulfuric acid potentiated the effect of bronchoconstrictor on airway conductance and flow rates at the lowest dose tested, 100 $\mu g/m^3$. In this test, the irritant potency more or less followed the acidity of the sulfates ($H_2SO_4 > NH_4HSO_4 > (NH_4)_2SO_4$). This is somewhat different than the irritant potential ranking indicated by guinea pig experiments discussed above.

The positive results of Utell et al. (1981) are in obvious contrast to the negative findings of other investigators at similar levels (Sackner et al., 1978; Avol et al., 1979). The authors offer four possible explanations:

1) Utell's group selected asthmatics according to carbachol reactivity; they should have been more homogeneous in terms of airway reactivity and, as a group, they may have been inherently more sensitive than other asthmatics tested;

2) Unlike the Avol subjects, Utell's subjects breathed orally;

3) Sackner's group studied asthmatics using daily medication to control bronchospasm. This medication may have reduced any response; none of Utell's subjects used medication; and

4) The parameters used by Utell may have been more sensitive to respiratory changes than those of the other investigators.

Of the above explanations, 1, 3, and 4 appear most plausible. Like Utell, Sackner employed oral only exposure, making the second point less pertinent. Perhaps a more important difference between Avol and Utell is that the former did not test bronchial reactivity. There was some suggestion of airway resistance changes in two of Avol's asthmatics. Sackner reportedly generated very small particles (≈ 0.1-0.2 μm) compared to the 0.5-1.0 μm particles of Utell. Considering hygroscopic growth in the respiratory tract, deposition of the larger aerosols on the sensitive receptors in the tracheobronchial region could have been substantially greater for similar initial concentrations. Moreover, the smaller aerosols used by Sackner would undergo more rapid neutralization by respiratory tract ammonia (Larson _et al_., 1977). The aerosols used by Utell should have more closely matched typical ambient size distributions.

The greater effectiveness of larger particle sizes (as well as longer exposure durations) may also be responsible for the increased respiratory symptoms reported by Horvath _et al._ (1981). Subjects were exposed for 2 hours to 0.9 μm sulfuric acid aerosol. Reports of sore throat, irritation and dryness increased at 220 $\mu g/m^3$ (3 of 9 subjects), again at 420 $\mu g/m^3$ (5 of 9 subjects), and at 940 $\mu g/m^3$ (8 of 9 subjects). Reports of cough paralleled those for the other symptoms. Other investigators reporting equivocal results or an absence of symptoms usually used smaller particle sizes and/or substantially shorter exposure periods.

A recent study on influenza patients indicates that bronchial reactivity to another soluble [illegible], sodium nitrate, ($NaNO_3$) is increased during infectious disease. In this study, Utell _et al_. (1980) examined influenza patients exposed to aerosols of $NaNO_3$ or NaCl (7,000 $\mu g/m^3$, ∿ 0.5 μm MMAD, $\sigma_g = 1.7$). Patients were exposed to both aerosols at the onset of sickness and at 1, 3, and 6 weeks after the initial exposure. Alternate exposures of NaCl or $NaNO_3$ lasted 16 minutes and were separated by 3 hours. Specific airway conductance and maximum expiratory flow at 60% of total lung capacity were significantly diminished following $NaNO_3$ exposure (but not NaCl) at the onset of sickness and exposure one week later, but no differences betweeen NaCl and $NaNO_3$ exposure treatments could be resolved after 3 weeks. The authors suggest several possible explanations for the increased sensitivity of the influenza patients. For example, epithelial damage exposing and sensitizing reflex receptors is postulated to result from viral infection.

Long term animal studies of effects of sulfuric acid or other soluble aerosols have produced either no lasting mechanical response or the reported respiratory functional measurements are of less interest than the results of direct examination of the damaged lung tissues. These studies are discussed in Section B.4.

2. Insoluble "Inert" Aerosols

Few animal studies have examined the effect of non-sulfur containing particles at levels approaching those observed in the ambient air. Studies in Amdur's laboratory of guinea pig exposures to fine oil mists, (∿ < 2.6 μm AED) spectrographic and activated carbon (< 1 μm CMD), MnO_2 (< .5 μm AED), open hearth dust (2 μm AED), and iron oxide (0.3 μm AED) have only found changes at relatively high concentrations (Amdur and Underhill, 1968, 1970; Costa and Amdur, 1979)*. These non-hygroscopic fine substances were apparently less irritating than sulfuric acid in this animal model.

Most of the controlled human exposure work involving insoluble particles was conducted before 1970. Using a variety of particle types and sizes (< .5 to over 12 μm), investigators have studied effects on adult workers and other subjects of high concentrations (2 to over 50 mg/m^3) for very short (2-10 minutes) to moderate (4-5 hours) duration. Even with differences among studies, the following findings appear to be relatively consistent:

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*Aerodynamic Equivalent Diameters (AED) reported here were calculated based on reported geometric count mean diameters (CMD). For these studies CMD's were < 1 μm.

1) Although there is variability among individual subjects, changes in pulmonary mechanics are concentration-dependent (Dubois and Dautrebande, 1958; McDermott, 1962; McKerrow, 1964).
2) Changes in pulmonary mechanics are most pronounced during the first few minutes and generally appear to diminish with time (Dubois and Dautrebande, 1958; McDermott, 1962; McKerrow, 1964).
3) The relative irritant potential of the various particulate substances (coal dust, carbon, calcium carbonate, iron hydroxide, india ink, charcoal and carbon) is difficult to compare due to differences in techniques, subjects, and exposures regimes.
4) The effects in polydisperse aerosol studies cannot be ascribed to fine particles alone. McDermott (1962) and McKerrow (1964) studied the effects of coal dust (9-50 mg/m^3, size < 7 μm), found increases in airway resistance following a 4 hour exposure and concluded that "significant increases in airway resistance occurred and the response was correlated to the weight of coarse particles between 3.6 and 7 μm." The lowest level tested in these healthy at rest subjects (9 mg/m^3) produced no significant responses, but all higher levels tested ($\geq$ 19 mg/m^3) did.

5) Symptomatic effects (tightness of the chest, dyspnea, cough and wheeze) may be observed concurrently even when broncoconstriction is clearly insignificant (Dubois and Dautreband, 1958). Similar studies comparing normal subjects and subjects with chronic lung disease (asthma, bronchitis, and emphysema) indicate that relative changes in respiratory mechanics can be similar in various diseased and normal groups following exposure to insoluble dusts. Some investigators have suggested that because many individuals with chronic lung disease have compromised pulmonary mechanics, the bronchoconstriction produced by particle exposure may place serious stress on such individuals at levels that should be easily tolerated in normal subjects (Constantine _et al_., 1959; Norris and Bishop, 1966).

A more recent study indicates that exposure to 2 mg/m^3 of "inert" polydispersed plastic dust containing carbon black (MMD was 9-12 μm, 30% mass > 12.5 μm) decreases forced expiratory volume at 1 second and forced expiratory flow in normal resting subjects (Andersen _et al_., 1979). Unlike the change in pulmonary mechanics, discomfort was proportional to the concentration of dust and was not immediate, lagging almost 2 hours behind changes in dust concentration. While the symptomatic effects may have caused personal discomfort, the noted results do not appear to be of substantial health import for resting normal healthy adults after 2-4 hour exposures. Exercise, oronasal breathing, or the presence of illness would increase the possibility of more significant responses for short term peak levels of such particles, perhaps at lower levels.

3. Particles and Pollutant Gases

The interaction of particles and gases has been reported to produce responses on respiratory mechanics and symptoms sometimes exceeding those attributable to the two agents administered separately. Enhanced responses may arise from deposition and combined action of gases and particles in various respiratory sites, physical sorption of gases on particles permitting enhanced respiratory tract penetration of the gas, and chemical reactions to form more irritant pollutants. The combination of SO_2 and particles has been most widely studied.

Based on the work of McJilton et al. (1976) and Snell and Luchsinger (1969), water or salt droplet aerosols may potentiate the effect of SO_2 by increased penetration, but probably not by oxidation to sulfuric acid. Amdur's work suggests the possibility that chemical transformation may occur within exposure chambers to enhance irritant potential of combinations of SO_2 and soluble, but not insoluble, aerosols (Amdur and Underhill, 1968). In some cases, stable sulfite-particle complexes might be implicated (Hansen et al., 1974); in others, sulfuric acid may be formed. Andersen et al. (1981) examined the combination of the dry "inert" carbon containing plastic dust described previously and sulfur dioxide. Effects were at most, additive. These combinations will be discussed more fully in the sulfur oxides staff paper.

Various particles also may enhance penetration and effects of other soluble gases, notably formaldehyde (LaBelle et al., 1955), and presumably, may increase effects on respiratory mechanics and symptoms.

4. Community Air Pollution

Changes in respiratory mechanics, or lung function, provide a sensitive and objective index of respiratory health status and have been employed in combination with respiratory symptom questionnaires to assess long-term alterations in populations exposed to air pollution. A number of community epidemiological studies have attempted to demonstrate an association between long-term exposure to particulate matter and impairments in respiratory mechanics and symptoms.

Table B-3 summarizes studies derived from the criteria document that best describe this relationship and are at minimum judged to be useful for qualitative comparisons. A few studies failed to show an association or show only a marginal one; they appear to compare populations with similar exposures to relatively low levels of particles. In general, populations living in areas with higher particulate pollution tend to have a higher prevalence of respiratory symptoms and lower lung function capability compared with other groups in areas with less particulate pollution. Many of the studies suffer from the design or methodological flaws associated with most cross sectional epidemiological studies (CD, p. 14-2 to 4). In addition, these studies rely on lung function tests that use devices and techniques subject to various methodological errors (e.g., calibration, seasonal effects, technician). Table B-3 indicates where these deficiences further limit a study's utility or reliability. In most studies, high levels of other pollutants, notably SO_2, were also present. These and other considerations, outlined in Chapter 14 of the criteria document, also apply to the epidemiological studies discussed in Sections B-F that follow.

In summarizing the studies of long-term (chronic) effects of particles and related pollutants on respiratory mechanics and symptoms, a clear and

TABLE B-3. EVIDENCE FROM EPIDEMIOLOGICAL STUDIES SHOWING QUALITATIVE ASSOCIATIONS BETWEEN CHANGES IN RESPIRATORY MECHANICS AND SYMPTOMS AND EXPOSURE TO PARTICLES

General Exposure Characteristics	Observed Effects/ Comments	Reference
Chronic Studies		
Children exposed to gradient of pollution within British industrial city whose pollution mix dominated by emissions from steel works and domestic coal combustion. After period of improving air quality, contrasts in pollution between areas narrowed by time of follow-up. Cross-sectional studies.	Progressively lower pulmonary function and progressively higher prevalence of upper and lower respiratory disease with increasing pollution exposure areas. Because of localized industrial regions and large changes in altitude within city, particle composition may have varied between areas. Health differences between areas not significant by time of follow-up study. Adequate controls. Aerometry problems (smoke calibration).	Lunn et al., 1967, 1970
Children living in 3 communities in Poland: Crakow, an industrial city (steel works, chemical, pharmaceutical and rubber factories) subject to long periods of fog and high levels of PM (BS) and SO_2; and two rural towns with low levels of pollution. Cross-sectional study.	Pulmonary function lowest in Crakow, highest in least polluted town. Bronchitis history, chronic cough, runny nose more prevalent in Crakow. Marginal differences between communities in prevalence of wheezing, and pneumonia. No differences in nonchronic cough and pneumonia. Controls appear adequate, but aerometry problems (smoke calibration).	Rudnik, 1978
Children and their families living in a Yugoslavian city (Zagreb), with relatively high levels and frequent peaks of PM ("suspended particulate matter") and SO_2, and in a nearby town with low levels of pollution. Cross-sectional study.	Pulmonary function among children lower in Zagreb and on days of higher pollution levels. Acute respiratory illnesses and lower respiratory tract illnesses more frequent in Zagreb among children, their mothers and siblings. Upper respiratory illnesses slightly more frequent among children in cleaner towns, but less frequent among their families. Limited pulmonary function testing, methods not described. Many variables measured, but no multivariate analysis. Monitoring sites not specified, long-term aerometric data provided.	Saric et al., 1981
Children living in copper smelter towns and other non-urban communities in Arizona. Smelter towns had relatively high levels of PM (TSP), characterized by stack emissions and fugitive dust. SO_2 levels among the communities varied. Cross-sectional study.	Lower pulmonary function among children living in cities with high TSP. No significant differences in rates of asthma, or respiratory symptoms. No association between SO_2 and pulmonary function or symptoms was found. Monitoring sites not specified. Limited control for socio-economic status.	Dodge et al., 1980
Children and parents exposed to contrasting high levels of PM (BS) and SO_2 in two London suburbs with domestic coal burning (1955-65). Surveys between 1962 and 1965 when smoke levels decreased and area differences disappeared. Cross-sectional	Higher prevalence of winter cough and phlegm among children and mothers (except in ex-smokers) in previously more polluted suburb compared with those in "cleaner" suburb. No area differences in pulmonary function among all subjects. Smoking, social class controlled. Symptom differences likely a residual effect of previously high pollution. Retrospective aerometry an important limitation.	Holland et al., 1969

TABLE B-3. EVIDENCE FROM EPIDEMIOLOGICAL STUDIES SHOWING QUALITATIVE ASSOCIATIONS BETWEEN CHANGES IN RESPIRATORY MECHANICS AND SYMPTOMS AND EXPOSURE TO PARTICLES (CONTINUED)

General Exposure Characteristics	Observed Effects/ Comments	Reference
Population of small town in northern New Hampshire (Berlin) was followed for 12 years. Town's major industrial emissions were from pulpmill, and were reduced in 1967, as indicated by lower PM (TSP and dustfall) and SO_x (sulfation rate) levels. In 1970, TSP declined to low levels; SO_x and SO_2 levels rose slightly to "moderate" levels. Cross-sectional study (1961) Two follow-up longitudinal studies (1967, 1970)	No difference in respiratory symptoms and lung function between different areas of town with different pollution levels in 1961. Incomplete aerometry; only 2 months of data. In first follow-up, after reductions in pollution levels, small improvements in lung function (peak expiratory flow rate, vital capacity) but little change in forced expiratory volume in one second. Prevalence of respiratory illnesses, e.g. chest colds, decreased. In second follow-up, no change in lung function or prevalence of respiratory illnesses and symptoms compared with previous study. Adequate controls. Aerometry limited by use of sulfation rates but SO_2 levels likely too low to be significant	Ferris and Anderson, 1962 Ferris et al., 1973; 1976
Comparison of Post Office employees, aged 40-59 yrs., in London, with high PM (dustfall) and SO_x (sulfation rate) pollution, and a cleaner rural town with lower level of domestic coal burning. Cross-sectional study.	Particularly over the age of 50, the London smokers had more and severer respiratory symptoms, produced more sputum, and had significantly lower average ventilatory function. Adequate controls. Imprecise aerometry, although differences in pollution between areas was clear.	Holland and Reid, 1965
Comparison of adults (25-74 yrs. of age) and children (6-8 yrs.) living in 6 U.S. cities of contrasting air pollution characteristics and various PM (TSP, MRP*, SO_4) and SO_2 levels. Cross-sectional phase of longitudinal study. *MRP = mass respirable particles	Among women, and male non-smokers, lung function was slightly lower in cities with highest PM levels. Male and female ex-smokers showed similar trend for wheezing, and male non-smokers reported a higher prevalence of phlegm in the "dirtier" cities. Trend towards higher prevalence of bronchitis in children in "dirtier" cities. No socio-economic controls. Ongoing study designed to show changes within population over time.	Ferris et al., 1979, 1980
Adolescents and adults living in Connecticut urban and rural towns with varying exposures to urban PM (TSP) and SO_2. Cross-sectional study.	No area differences in respiratory mechanics or prevalence of chronic bronchitis. Prevalence of upper respiratory symptoms (e.g. cough, phlegm, dyspnea) increased from lifetime rural residents to mixed urban/rural residents to lifetime urban residents. Asthma more common in rural area. Adequate controls. Limited aerometry data provided on previous exposures. Mixed results make interpretation difficult.	Bouhuys et al., 1978
Acute Studies		
Exercising children, adolescents and adults in Tuscon, Arizona exposed to moderate peak daily levels of PM (TSP) and low SO_x levels.	Pulmonary function decreased after exercise on days of moderate TSP levels and high temperature. Effects greater among children and adolescents than in adults. Effects of temperature and pollution not separated.	Lebowitz et al., 1974
Children living in Steubenville, Ohio exposed to high air pollution episodes in which PM (TSP) and SO_2 reached high daily levels.	Relatively small declines in pulmonary function one day after episodic levels of pollutants. Effects persisted at least one-three weeks. Suggestive but inconclusive results due to preliminary analyses. Good exposure estimates.	Dockery et al., 1981

consistent association emerges for higher exposures that applies to children as well as adults. Long-term occupational exposures to various particles at high concentrations also have been implicated in the prevalence of airway obstruction, as indicated in Table B-1. These findings qualitatively support the evidence from community studies which indicate that an association exists between respiratory mechanical impairments and long-term exposures to elevated particle levels.

In contrast, the information on acute effects of ambient particles on respiratory mechanics is limited to two studies, also summarized in Table B-3. Lebowitz _et al_. (1974) suggests that the observed pulmonary function decrements in exercising children and adolescents exposed to southwest U.S. urban aerosols may be linked to short-term exposures to particles at high temperature and that the mechanism for chronic effects might be mediated by acute insults to the respiratory tract. Dockery _et al_. (1981) reported on preliminary studies of acute respiratory mechanical changes in children exposed to higher concentrations of particulate matter in Steubenville. The investigators found that children exposed to sudden increases of TSP and SO_2 concentrations had declines of 1 to 2% in forced expiratory volume in 1-second ($FEV_{1.0}$). These changes were considered small relative to sampling variability and marginally significant. The authors concluded that this study provides suggestive but inconclusive evidence concerning the relationship between short-term changes in air pollution concentration and the level of $FEV_{1.0}$. The long-term significance of these effects remains to be determined.

A somewhat unique controlled study of community air pollution also supports these acute effects associations. Toyama (1964) exposed 10 healthy males to high levels (10 mg/m^3) of resuspended dust collected by deposition in a Japanese urban area. Exposures were brief, consisting of 20 deep breaths. The dust ranged from 0.5 to 10 μm (MMD ∿ 2.0 μm) and was composed of crustal material (64% as SiO_2, Al_3O_2, CaO, MgO), sulfates (10%), and volatile material (19%). Bronchoconstriction effects as measured by airway resistance varied with individual, ranging from no response (one subject) to a 70% increase. The response appeared to be mediated by a reflex reaction to mechanical irritation and was said by the authors to be consistent with insoluble dust studies (see Subsection B.1.b. above). Subsequent experiments suggested a greater than additive effect of resuspended dust and SO_2 (> 3 ppm).

B. Aggravation of Existing Respiratory and Cardiovascular Disease

Because bronchoconstriction is one of the major mechanisms by which particles may aggravate existing respiratory disease, many of the animal and human studies most relevant to this issue have been discussed in the preceeding section; this section will focus on the results of community epidemiological studies.

The epidemiological evidence suggesting an association between particles and aggravation of the various states of respiratory illness and associated cardiovascular disease is summarized in Table B-4.

Evidence from the "smog" episodes of Donora, London, and New York City, in the 1950-60's indicate that individuals with pre-existing respiratory and cardiac illness are especially vulnerable to sudden increases in air pollution. Furthermore, the acute changes or aggravations of their conditions provide a more sensitive index of the effects of air pollution than do day-to-day changes in relatively healthy individuals.

TABLE B-4. EVIDENCE FROM EPIDEMIOLOGICAL STUDIES SHOWING QUALITATIVE ASSOCIATION BETWEEN AGGRAVATION OF PRE-EXISTING CARDIO-RESPIRATORY DISEASE AND EXPOSURE TO PARTICLES

General Pollution Characteristics	Observed Effects/Comments	Reference
Severe air pollution episodes lasting several days. Donora, London episodes characterized by dense fog, prolonged continuous exposures. NYC episodes had less fog, more diurnal variation in pollution. Complex pollution mix dominated by emissions from fossil fuel combustion, industrial processes, other urban sources and characterized by sudden increases in PM and SO_2 to extremely high levels.	Increased illness (and/or mortality), especially among individuals with pre-existing cardio-respiratory disease (e.g. bronchitis, emphysema, cardiopulmonary disorders). Difficult to separate effect of PM from SO_2, weather variables, and infection.	Schrenk et al., 1949 Ministry of Health, 1954 Greenburg et al., 1963; Glasser et al., 1967; Becker et al., 1968
Follow-up of Donora, Pa. survivors	Survivors of Donora episode with prior chronic disease, and those who became acutely ill during episode had higher subsequent mortality and illness.	Ciocco and Thompson, 1961
Minor pollution episodes during two London winters when emissions from doemstic coal combustion were high, characterized by periodic increases in PM (BS) and SO_2 to high levels.	Apparent increase in hospital admissions (and mortality) above 15-day moving average, especially among individuals with pre-existing cardio-respiratory disease. Temperature, humidity were accounted for in the analyses.	Martin, 1964
Daily fluctuations in Steubenville, Ohio pollution mix characterized by PM (TSP), SO_2, and NO_2 that periodically reached high levels.	Quartile analysis shows hospital emergency room visits for cardio-respiratory diseases, all disease categories, not significantly associated with pollution levels; small increase at the high levels. Linear regression showed small but statistically significant effects of TSP and SO_2, but not other pollutants. Controls for weather, temporal factors.	Samet et al., 1981
Daily fluctuations in Canadian industrial city pollution mix dominated by emissions from steel production operations, and characterized by PM (TSP) and SO_2.	Weekly hospital admissions for acute respiratory disease (e.g. bronchitis, emphysema, pneumonia, asthma) associated with PM and SO_2, and not other pollutants. No direct adjustments for season. Adjustments for temperature provides indirect control.	Levy et al., 1977
Daily and hourly fluctuations in London air pollution mix dominated by emissions from domestic coal combustion and characterized by concentrations of PM, (BS), SO_2, and often sulfuric acid, that reached high short-term levels.	Exacerbation of condition among patients with severe bronchitis and emphysema associated with daily elevations of pollution. Authors suggest that effects more likely caused by brief concentration peaks (few hours). Good aerometry; weather variables controlled.	Lawther et al., 1958, 1970
Fluctuations in urban location (U.S.) pollution dominated by emissions from grain storage operations, other industrial emissions and characterized by periodic increases in PM (CoH) and low SO_2.	Asthmatic attacks among college students associated with organic dusts and incomplete combustion products. Composition analysis of CoH filters. No correlation between TSP or sulfate particulates and asthma. Weather variables controlled.	Smith and Paulus, 1971
Fluctuations in urban location (U.S.) pollution mix containing emissions from industrial processing and combustion, fossil fuel combustion, and vehicular traffic, and characterized by periodic high particulate (TSP or CoH) and SO_2 levels.	Increased visits to hospital for asthma on days of high air pollution. No specific pollutant can be incriminated.	Glasser et al., 1967 Girsh et al., 1967
	No association found between NYC hospital emergency admissions for asthma and smokeshade. Single monitoring station used to estimate city-wide exposure (Greenburg). No temperature adjustment (Goldstein and Dulberg).	Greenburg et al., 1967 Goldstein and Block, 1974 Goldstein and Dulberg, 1981

Based on the possible responses to particle deposition in the tracheobronchial and alveolar regions as outlined in Section V.A, and the susceptibility of cardiopulmonary patients to stress from infections or increased airway obstruction (Fishman, 1976), the observed associations are biologically plausible.

Effects of ambient particles on asthmatics are also to be expected. Asthma is a disease characterized by a high degree of airway reactivity, with a plurality of causes (allergy, inflammation, irritation, temperature changes, etc.) and a wide variation of physical manifestations. These factors make asthmatics an important and difficult group to study. Despite these difficulties, and the limitations in assessing personal exposures inherent in epidemiological research, some studies have attempted to delineate relationships between aggravation of asthma and particulate matter. Although the results are equivocal, there is an indication that an association may exist, especially when the particle pollution contains dusts derived from biologic matter (e.g., grain dust). Due to their apparent sensitivity to SO_2 (Sheppard et al., 1981; Koenig et al., 1981) epidemiological studies of asthmatics where high SO_2 levels were present (e.g., Cohen et al., 1972), cannot be used as qualitative support for an association with particulate matter.

It must be emphasized that in most of the studies of acute exacerbations of chronic or acute respiratory conditions, the effect of particles cannot be distinguished from other pollutants, usually SO_2. However, clear evidence emerges from the body of epidemiological literature that implicates particles in aggravating disease among bronchitics, asthmatics, cardiovascular patients and people with influenza.

C. Alterations in Host Defense Mechanisms--Clearance and Infection

1. Clearance

The effects of particulate exposure on clearance have been examined using several different types of particles. Studies of exposures to cigarette smoke and sulfuric acid have received more attention than other particulate types. These studies have focused on mucociliary clearance in various regions of the respiratory tract.

As indicated in Table B-5, the exposure-dependent response of mucociliary clearance rates to particle exposure can be complex. Observations in humans indicate that a single exposure to as little as 100 $\mu g/m^3$ of sulfuric acid for 1 hour can accelerate bronchial clearance rates in some individuals. Leikauf et al. (1981) note that as sulfuric acid concentrations are increased, mucociliary clearance rates are progressively reduced. Although donkeys exposed by the same investigators do not appear to show any change in mucociliary clearance rates below about 200 $\mu g/m^3$ sulfuric acid (1 hour), depressions in mucociliary clearance rates have been reported in donkeys following repeated exposures of 100 $\mu g/m^3$ sulfuric acid for repeated 1 hour exposures (5 days/week for 6 months). Two (of four) donkeys in this experiment developed grossly abnormal clearance times during the fourth through sixth months of exposure and remained so for at least three months after exposure was ended (Schlesinger et al., 1979).

Lippmann and co-workers (1981) have stressed several similarities between the above effects of sulfuric acid on clearance and those of cigarette smoke: 1) transient accelerations of clearance are produced by low dose exposures (100 $\mu g/m^3$ sulfuric acid for 1 hour or 2 to 7 cigarettes); 2) a transient slowing of clearance follows high dose exposure (1 mg/m^3 sulfuric acid for 1 hour or $\geq$ 15 cigarettes); and 3) alterations in clearance rates

TABLE B-5. EFFECTS OF SULFURIC ACID, CARBON EXPOSURE ON MUCOCILIARY CLEARANCE

Clearance Region	Pollutant Exposure	Time After Exposure	Effect(s)	Comment	Reference
Tracheobronchial	Sulfuric Acid Ten healthy adults exposed for one hour (via nasal face mask) to: a) 110 $\mu g/m^3$ H_2SO_4 (0.5 μm MMAD, $\sigma_g = 1.9$) b) 330 $\mu g/m^3$ c) 980 $\mu g/m^3$	0-10 hours	a) Acceleration in bronchial mucociliary clearance	Six of ten healthy non-smoking adults exhibited increases in bronchial clearance half times that were greater than 25% of control values. As a group, a significant ($P<0.02$) 40% decrease in mean bronchial clearance half times was observed.	Leikauf et al., 1981
			b) Varied	Three persons showing substantially faster clearance than in the control test and three persons showing substantially slower clearance than in the control test.	
			c) Depression in bronchial mucociliary clearance rates	As a group, the mean rate of clearance was significantly depressed ($P<0.03$) with mean bronchial clearance half times being about 150% of control values.	
Tracheobronchial	Four donkeys exposed for 1 hour (via nasal catheters) to: 194 $\mu g/m^3$ H_2SO_4 (0.3 - 0.6 μm MMAD, $\sigma_g = 1.5$)	0-24 hours	Depression in bronchial mucociliary clearance	Transient increase in bronchial clearance half time in one of four donkeys. Transient increases in two other animals were observed at higher concentrations. Persistent alterations in clearance produced in two animals.	Schlesinger et al., 1978
Trachea	Eight dogs exposed for 1 hour (via nasal face mask) to: 500 $\mu g/m^3$ H_2SO_4 (0.9 μm MMAD, $\sigma_g = 1.3$)	a) 30 minutes	a) No group change	One dog (#6) showed an increase in tracheal mucus velocities of about 300% of control values at 30 minutes after exposure.	Wolff et al., 1981
		b) 1 week	b) Depression in tracheal mucus flow rate	A significant depression ($P<0.05$) was observed for the group. One dog (#6) showed an increase in tracheal mucus flow rate unlike the rest of the group.	
Tracheobronchial	Four donkeys exposed for 1 hour per day; 5 days per week; for 6 months-122 exposures (via nasal catheters) to: 105 $\mu g/m^3$ H_2SO_4	a) 1 week	Bronchial clearance becomes erratic	The bronchial clearance half times became significantly different from controls on many test days.	Schlesinger et al., 1979
		b) 6-9 mos.	Varied	Two animals sustained impairment of clearance towards the end of the 6-month exposure period and continued to have erratic clearance during a 3-month follow-up period.	
Trachea	Hamsters exposed for 3 hours to: 1.1 mg/m^3 H_2SO_4 (0.12 μm) and/or 1.5 mg/m^3 carbon (0.3 μm)	Immediate	Ciliary beat frequency depressed by both H_2SO_4 exposures	Morphological damage was greatest for the H_2SO_4 and carbon exposure	Schiff et al., 1979

that can persist for several months following multiple exposures (> 6 one hour exposures at 200-1000 $\mu g/m^3$ sulfuric acid, 6 months of daily exposure to 100 $\mu g/m^3$ sulfuric acid for 1 hour/day, and/or 4 to 8 months of 30 cigarettes for 3 times/week).

Based on these similarities, together with the well established role of cigarette smoking in the development of chronic bronchitis in humans, it is suggested that sulfuric acid exposure may play a role in the etiology of chronic bronchitis (Lippmann et al., 1981). Significantly, although these sulfuric acid levels are perhaps a factor of two or more of peak U.S. levels, they are in the range of peak levels of sulfuric acid during London episodes as reported by Commins and Waller (1967). Even in London, however, such levels apparently were rare.

Several clearance studies have also been conducted using carbon or carbon/SO_2 mixtures. Camner et al. (1973) reported that brief but extremely high exposure to 11 μm carbon particles usually accelerated clearance. In contrast to the studies on tracheobronchial clearance, normal subjects have not shown changes in nasal mucous clearance rates following exposure to polymerized plastic dust containing carbon black below 25 mg/m^3 (Andersen et al., 1979).

2. Infection

As indicated above, changes in mucociliary clearance are the most studied effects on host defense systems. No controlled human exposure studies have evaluated the effect of particulate exposure on other mechanisms affecting susceptibility to infection. Table B-6 lists animal studies highlighted in the criteria document. Animal exposure studies indicate that single or repeated exposures to high levels of toxic metal salts increases

TABLE B-6. EFFECT OF PARTICULATE MATTER ON HOST DEFENSE SYSTEMS

Concentration	Observation	Reference
Acute exposures; Infectivity Model		
Mice exposed for 2 hours to: 0.075 - 1.94 mg/m^3 $CdCl_2$ (94-99% < 1.4 µm)	Increased mortality at .1 mg/m^3 Cd following bacterial challenge (*Streptococcus pyogenes*)	Gardner *et al.*, 1977b
0.1 - 0.67 mg/m^3 $NiCl_2$ (94-99% < 1.4 µm)	Increased mortality at 0.5 mg/m^3 Ni following bacterial challenge (*S. pyogenes*)	Adkins *et al.*, 1979
0.5 - 5 mg/m^3 Mn_3O_4 (94-99% < 1.4 µm)	Increased mortality at 1.55 mg/m^3 Mn following bacterial challenge (*S. pyogenes*)	Adkins *et al.*, 1980
Mice exposed for 3 hours to: 0.1 ppm O_3 followed by 2 hours at 0.9 mg/m^3 H_2SO_4 (0.23 µm AED, $\sigma_g = 2.4$)	Increased susceptibility to infection by *S. pyogenes.* Neither pollutant alone produced an effect. Order of exposure (ozone first) was important.	Gardner *et al.*, 1977a
Chronic Exposures		
Mice exposed for 100 hours per week for 192 days to: 0.56 mg/m^3 carbon (1.8 to 2.2 µm MMD) and/or 5.24 mg/m^3 (2 ppm) SO_2	Immunological (and morphological) alterations in the lungs. Systemic immune system affected after 192 days. SO_2 + carbon and carbon alone were more effective than SO_2 alone.	Zarkower, 1972
Mice exposed for 3 hours per day for 5 days per week for 20 weeks to: 1.4 mg/m^3 H_2SO_4 (0.12 µm/m^3) and/or 1.5 mg/m^3 carbon (0.3 µm/m^3) or 2.9 mg/m^3 H_2SO_4 coated carbon (0.4 µm).	Increased mortality and pulmonary consolidation following exposure to virus in the acid coated carbon. No change in resistance to *Klebsiella pneumoniae* (a bacteria). Immunological changes were similar to those observed by Zarkower, 1972.	Fenters *et al.*, 1979

the rate of mortality resulting from induced bacterial infection (Maigetter et al., 1976; Gardner et al., 1977b; Adkins et al., 1979, 1980). Using the same model, however, no increased mortality rates were observed in animals exposed to sulfuric acid, ammonium sulfate, iron oxide, and carbon black (Gardner et al., 1977a; Gardner, 1981; Ehrlich et al., 1978; Ehrlich, 1979). Sulfuric acid and ozone, however, increased mortality even though neither pollutant alone produced an effect. Long-term studies of combinations suggest that high levels of carbon or carbon plus SO_2 or sulfuric acid may affect the immune system (Zarkower, 1972; Fenters et al., 1979).

The mouse studies in Table B-6 suggest that high levels of particles and gas (O_3, SO_2)--particle combinations may increase susceptibility to a bacterial or viral challenge and produce immunological changes. The relevance of many of these high dose results to real world exposures can, however, be questioned. Of similar interest are in vitro and in vivo studies of various particles on alveolar macrophages which are the primary defense cells of the lungs. A variety of trace elements, minerals, fibrous materials, and other particles have been observed to damage macrophages to varying degrees (CD, Section 12.3.4.2).

3. Clearance and Infection - Community Air Pollution

The first part of Table B-7 summarizes the studies that demonstrate the influence of particulate pollution on acute infections in populations.

Insults to the respiratory tract such as acute infections in childhood may be important in the subsequent development of obstructive airways disease, where exposure to adverse environmental factors changes a relatively minor infection into one that produces permanent changes in the respiratory tract (Colley et al., 1973; Kiernan et al., 1976).

Because the impact of ambient pollutants on respiratory illness may have important consequences for the subsequent development of functional abnormalities, and a substantial proportion of these illnesses in very young children involve the lower respiratory tract (Speizer et al., 1980) and particularly the small airways (Tager and Speizer, 1976), the studies in the second part of Table B-7 relating chronic exposures to pollution to childhood respiratory infection are especially significant.

Based on animal and clinical data, episodic infection with a variety of microbial agents might be an important risk factor in the evolution of mucus hypersecretion and chronic obstructive airways disease. (Speizer and Tager, 1979). It is thought that upon irritation, excessive mucus is secreted into the airways, causing airway blockage and reduced mucus clearance. This damage predisposes the individual to infection, particularly in the lower airways. At this stage, characterized by cough and phlegm production, the effect may be reversible. Chronic irritation, however, may allow infections to persist in the airways, ultimately producing irreversible obstruction. Thus, impaired mucus clearance appears to be an important indicator in the evolution of obstructive diseases such as bronchitis.

Studies of normal population groups, with comparable smoking and age distributions suggest a greater prevalence of symptoms of mucus hypersecretion (e.g., runny nose, persistent cough, phlegm production) in subjects living in more polluted environments (Holland and Reid, 1965; Holland et al., 1969; Lambert and Reid, 1970; Rudnik et al., 1978). To the extent that the longitudinal study of Fletcher et al. (1976) is related to air pollution, it distinguishes the effect of particles from SO_2 with regard

TABLE B-7. EVIDENCE FROM EPIDEMIOLOGICAL STUDIES SHOWING QUALITATIVE ASSOCIATIONS BETWEEN RESPIRATORY TRACT INFECTIONS/ALTERED CLEARANCE AND EXPOSURE TO PARTICLES

General Pollution Characteristics	Observed Effects/Comments	Reference
Acute Studies		
Periodic pollution episodes and fogs during a London winter marked by intensive domestic coal combustion.	Minor respiratory illness and infection (e.g. colds) rates correlated with weekly peaks of smoke and SO_2. No information on daily levels of pollution and corresponding health effects.	Angel et al., 1965
Severe pollution episode in NYC characterized by high PM (CoH) and SO_2 levels.	Increased upper respiratory infections in 4 old-age homes. No significant increase in daily mortality or in visits for respiratory or cardiac complaints in city hospitals and clinics. Inadequate aerometry (one monitoring station).	Greenburg et al., 1963
Contrasting levels of "dust" pollution and high levels of SO_2 and NO_2, during two influenza epidemics, 3 years apart in an industrial city in Bulgaria.	Significant correlation between "dust" pollution and SO_2 and incidence of influenza cases during epidemic in which dust levels were high, compared with second epidemic when correlation disappeared and dust levels were lower. Aerometry not described.	Kalpazanov et al., 1976
Daily fluctuations in NYC 1962-65 pollution mix as characterized by periodic increases of PM (CoH) and SO_2.	Significant correlations between acute respiratory symptoms (cough, colds) measured weekly, in adults and children, and pollution and meteorological variables. 1-3 day lag effects. Inconsistent associations with SO_2. Smoking controlled. Adequate aerometry but no reliable exposure levels.	McCarroll et al., 1967; Mountain et al., 1968; Thompson et al., 1970; Cassell et al., 1972
	Sensitive sub-group mostly children (< 10 yrs.) identified-cold symptions of greater duration and severity. Domestic crowding and smoking not controlled.	Lebowitz et al., 1972
Chronic Studies		
British children exposed to gradient of PM (BS) and SO_2 levels depending on degree of domestic coal combustion in area of residence.	Frequency of lower respiratory infections higher among children in higher pollution areas. No association between pollution and upper respiratory tract infection. At ages 20 and 25, prevalence of respiratory symptoms related primarily to smoking habits although some relationship with histories of respiratory illness persisted. No direct pollution measurements although estimates considered reasonable.	Douglas and Waller, 1966 Colley et al., 1973 Kiernan, et al., 1976

TABLE B-7. EVIDENCE FROM EPIDEMIOLOGICAL STUDIES SHOWING QUALITATIVE ASSOCIATIONS BETWEEN RESPIRATORY TRACT INFECTIONS/ALTERED CLEARANCE AND EXPOSURE TO PARTICLES (CONTINUED)

General Pollution Characteristics	Observed Effects/Comments	Reference
Gradient of PM (BS) and SO_2 exposures of children living in 4 areas of industrial city whose pollution mix dominated by emissions from steelworks and domestic coal combustion. Contrasts in pollution between areas narrowed by time of follow-up.	Upward gradient of upper and lower respiratory infection prevalence with increasing pollution exposure. Because of localized industrial regions and large changes in altitude within city, particle composition may have varied between areas. Health differences between areas not significant by time of follow-up. Adequate controls. Aerometry problems (smoke calibration).	Lunn et al., 1967; 1970
Children and their families living in a Yugoslavian city (Zagreb) with relatively high levels and frequent peaks of PM ("suspended particulate matter") and SO_2 and in a nearby town with low levels of pollution.	Acute respiratory illnesses and lower respiratory tract illnesses more frequent in Zagreb among children, their mothers and siblings. Upper respiratory illnesses slightly more frequent among children in cleaner town, but less frequent among their families. Pulmonary function among children lower in Zagreb. Limited health measurements, methods not described. Many variables measured, but no multivariate analysis. Monitoring sites not specified, long-term aerometric data provided.	Saric et al., 1981
Adults or children living in urban and rural areas and exposed to contrasting PM and SO_2 pollution because of higher emissions from fossil fuel combustion, industrial processes, and vehicular traffic in urban areas.	Subjects in more polluted areas had higher prevalence of mucus hypersecretion symptoms (e.g., runny nose, cough, phlegm). Controls for smoking and socio-economic status; latter not controlled in Lambert and Reid. Aerometry did not yield reliable exposure estimates. Areas had sufficiently different pollution characteristics to permit qualitative conclusions however.	Holland and Reid, 1965 Holland et al., 1969 Lambert and Reid, 1970 Rudnik et al., 1978
London men exposed to progressively lower PM (BS) levels, and stable SO_2 levels, during a period of domestic coal combustion control.	Unexpected decline in mean sputum volume with declining PM levels in men who had no change in smoking habits. Potential confounders include reductions in tar content of cigarettes during study period and unavoidable variability in sputum collection. These complicate interpretation. No reliable exposure estimates.	Fletcher et al., 1976

to impaired mucus clearance and is consistent with the evidence that suggests that particles, with and without SO_2, have acute and long-term impacts on the respiratory system that may increase its susceptibility to infection.

D. Morphological Damage

Damage to respiratory tract tissues has been associated with exposure to a variety of different types of particles, including direct acting irritants (sulfuric acid) and insoluble dusts. Because detection of such damage normally requires invasive techniques, evidence for these effects is derived from controlled animal tests and autopsy studies of individuals characterized as having long-term exposures to urban, rural, or occupationally-related particles. As such, evidence for morphological damage is mainly qualitative.

1. Sulfuric Acid/Combinations

Table B-8 summarizes a variety of studies examining the effects of sulfuric acid and fly ash alone or in combination with other pollutants. As discussed earlier, a serious complication of all animal acid aerosol exposure studies is the potential neutralization by chamber ammonia. In the absence of special precautions, chamber ammonia concentrations may average up to 25 ppm (Malanchuk et al., 1980). Thus, the sulfuric acid exposure studies listed may actually have examined ammonium sulfates/sulfuric acid mixtures, even where frequent cage cleanings may have reduced contamination. Nevertheless, some morphological effects have been observed in some studies.

The chronic exposure of beagle dogs provide several interesting findings (Hyde et al., 1978; Gillespie, 1980). Distinct, pathological changes in structure and pulmonary mechanics were found (by electron microscopy) at

TABLE B-8. RESULTS OF SELECTED LONG-TERM ANIMAL STUDIES OF EFFECTS OF LUNG STRUCTURE AND FUNCTION

Exposure	Observations	Reference
Beagle Dogs Exposed a) 21 hr/day, 620 days to 1) 13.4 mg/m³ SO_2 2) .76-.89 mg/m³ H_2SO_4 < .5 µm CMD 3) Combination of above (Conc. x time: SO_2 ≈240 mg/m³ - months H_2SO_4 ≈15 mg/m³ - months)	After 225 days Functional changes in all groups relative to controls, nature varied for 3 groups After 620 days More functional changes with H_2SO_4 No hematological changes Decreased lung and heart weight in H_2SO_4 exposed groups No morphological changes detected in any group No post exposure examination reported	Lewis *et al*., 1969, 1973
b) 16 hr/day, 68 months to 1.1 mg/m³ SO_2 + 0.09 mg/m³ H_2SO_4, <.5 µm CMD. (Conc. x time: SO_2 ≈50 mg/m³ - months H_2SO_4 ≈ 4 mg/m³ - months)	After 18-68 months No functional changes 18, 36 months Some changes at 61 months, end of exposure 24 months after end of exposure Functional losses worsened 32-36 months after end of exposure Increased lung weight, total lung capacity A number of morphological changes, including loss of cilia in conducting airways; possible bronchiolar cell hyperplasia; loss of interalveolar septa; enlarged air spaces; "striking" increase in number and site of interalveolar pores, emphysema	Hyde *et al*., 1978; Gillespie, 1980
Guinea Pigs, Monkeys Exposed 52-78 weeks to various combinations of SO_2 (0.1-5 ppm), H_2SO_4 (0.1-1 mg/m³, 0.5-3.4 µm MMD), Flyash (≅0.5 mg/m³; 3.5-5.9 µm MMD)	Morphological changes in bronchial mucosa (monkey) greatest for 1 ppm SO_2 + 0.93 mg/m³ H_2SO_4 with or without flyash, less for 0.99 H_2SO_4 + .55 mg/m flyash. None observed for other exposures or groups. Results indicate: a) H_2SO_4 (between 0.1-1 mg/m³ may produce morphological effects) SO_2 + H_2SO_4 results equivocal b) Flyash accumulated in lung, no obvious effects on bronchial morphology c) Flyash/SO_2 combinations produced no detected effects No post exposure examination reported	Alarie *et al*., 1973a, 1973b, 1975

exposure levels that were considerably lower than those used in earlier experiments. As discussed in the criteria document (CD, p. 12-60), the evidence suggests that pathological changes continued to become progressively more severe with time after exposure had been terminated, highlighting the importance of post-exposure follow-up studies. The authors consider the morphological changes to be analagous to an incipient stage of emphysema.

In applying these beagle dog studies to real world exposures, the following points should be considered:

1) The alternating exposure/clean air system was useful, but does not simulate repeated short-term peaks or lower long-term averages more likely to be experienced in the ambient air.

2) Based upon the information presented in the studies, it is difficult to determine whether the particle size was representative of that normally found in ambient air.

3) Actual sulfuric acid exposures were probably lower due to unknown chamber levels of endogenous ammonia. Neutralization is faster for smaller particles.

4) It seems reasonable to attribute most of the deep lung changes to the sulfuric acid or acid/ammonium sulfate mixture, based on the probability of deep lung deposition. Sulfuric acid aerosols are unlikely to absorb SO_2 and increase penetration, although ammonium sulfate aerosols might.

5) If the two sets of dog studies can be directly compared, they indicate that morphological effects of the SO_x mixture are more pronounced with extended lower level exposure. This is in contrast to acute bronchoconstrictive responses for these substances,

where higher concentrations appear more important than exposure duration. Although the time integrated dose (concentration x time) in the Hyde et al. study was lower than in the Lewis work, morphological effects were apparently much more pronounced, suggesting time integrated dose is not necessarily a good indicator of morphological damage.

The Alarie et al. (1975) studies also suggest that sulfuric acid (between 0.1-1 mg/m^3), may affect bronchial morphology; in two cases, sulfuric acid was more potent when combined with SO_2, in one case it was not. Fly ash accumulated in the lungs but apparently (using light microscopy) did not affect morphology, even in combination with SO_2. Although 3-5 μm fly ash can be efficiently deposited in mouth breathing humans, the alveolar penetration in guinea pigs and small monkeys might well be significantly lower. Pulmonary function testing suggested possible effects for most sulfuric acid combinations tested (0.1 - 1 mg/m^3), but the differences in response to these levels were not large and interpretation difficult due to anomalous trends in the control group. In view of the beagle dog studies, (Gillespie, 1980), it is unfortunate that post-exposure follow-up studies were not conducted.

2. Insoluble Particles

As indicated earlier, slowly cleared insoluble particles may accumulate in the alveolar region of the lung. Controlled animal exposures and in vitro studies have typically been conducted at unrealistically high concentrations and are most useful in identifying mechanisms. (NIOSH, 1974; White and Kuhn, 1980; Ziskind et al., 1976). Besides accumulation in lung tissue

TABLE B-9. RETROSPECTIVE PATHOLOGY STUDIES

Pollutant Mixture	Population	Observations	References
Granite Quarry	Granite Workers	Fibrotic lesions associated with deposits of crystalline silica were found postmortem in the lungs of 15 granite workers (a total of 15 workers were examined).	Craighead and Vallyathan, 1980
Ambient Air	Farm Workers/ Rural Resident	Abnormal numbers of birefringent particles have been found in the lungs of seven patients in association with a spectrum of early to late interstitial inflammation and fibrosis. Particles found in lungs were also sampled from soil samples.	Sherwin et al., 1979
Descrt Air	Desert Dwellers	Silica containing dust particles were distributed in the lymphatic bed in early stages of silicate pneumoconiosis and in fibrous scars in later involvement.	Bar-Ziv and Goldberg, 1974
Ambient Air	Humans	Observations of five human lung specimens indicated an accumulation of dust laden and crystal laden macrophages at the site of lymphatics and a mild fibrosis existed around these dust granulomas. No occupational or personal exposure could explain this pneumoconiosis and most likely the individuals accumulated the mica in their lungs from atmospheric air.	Brambilla et al., 1979
Ambient Air (Zoo)	65 Mammals and Marsupials	Accumulation of crystal-laden macrophages was the only lesion in about 50% of the cases. There was a significant focal thickening of septums while the surrounding pulmonary parenchyma was normal. In 5% of the animals a more severe fibrosis occurred with surrounding emphysema. The intensity of pneumoconiosis was greater in older animals.	Brambilla et al., 1979

and macrophages, insoluble particles (most notably various free silica minerals) may indirectly produce fibrosis through macrophage damage.

Autopsies of both humans and animals naturally exposed to various kinds and amounts of fugitive crustal material have found changes in many ways similar to the morphological responses produced in controlled studies (see Table B-9). The occurrence of silicosis in workers exposed to high levels of silica in occupations of sandblasting and granite mining is well documented (Ziskind et al., 1976; NIOSH; 1974). Evidence for varying degrees of penumoconiosis has recently begun to appear in the literature for animals and people exposed to crustal dust near or not far above ambient levels. Silicate pneumoconiosis has been reported in zoo animals (Brambilla et al., 1979), farm workers (Sherwin et al., 1979), desert dwellers (BarZiv and Goldberg, 1974), and residents of the southwestern United States (Brambilla et al., 1979).

Routine examination of over 11,000 San Diego zoo animals exposed to ambient particulate matter indicated the presence of crystalline material in 20% of the cases (Brambilla et al., 1979). Accumulation of crystal-laden macrophages was the only lesion in about 50% of the cases, but more serious lesions were observed in some animals. Macrophages and birefringent particles had accumulated in pulmonary lymphatics. Crystals from lung tissues were lamellar, 1-10 μm long and 0.5-7 μm wide, and were chemically and physically similar to those found in local air samples as determined by electron microscopy and X-ray analyses. The authors noted that they had observed similar accumulations and responses in autopsies of various human residents of this area, apparently not related to occupation.

Sherwin et al. (1979) found fibrotic lesions in 6 vineyard workers and one farm housewife. Particles measuring up to 3 by 12 μm were found in the lungs and were minerologically similar to local soil samples. The deaths of these subjects were apparently related to respiratory difficulties, but the role of high soil particle loadings cannot be unequivocally separated from possible pesticide contamination, organic dusts, and in some cases, smoking. Nevertheless, these coarse particles apparently produced responses in lung tissues, since in most cases interstitial deposits of particles were associated with areas of chronic inflammation and fibrosis.

A study of Bedouin desert dwellers identified several degrees of accumulation and response (see Table B-9) associated with exposure to desert dust of undetermined particle size (Bar-Ziv and Goldberg, 1974). Accumulation tended to increase with age and probable degree of exposure to higher concentrations. In some cases mild focal emphysema was present in areas surrounding dust nodules. The authors suggest this was not of clear clinical significance. A number of pathological conditions were noted in the 54 lung specimens examined, but effects were not necessarily related to dust accumulation. More advanced nodules characteristic of classical occupational silicosis were not observed. The authors suggest that the limited fibrotic response might be explained by the reduced activity of aged desert dust compared to freshly generated particles. Cigarette consumption or other personal particle exposure unrelated to desert dusts were not evaluated.

Although the findings are difficult to ascribe to particulate air pollution, Ishikawa et al. (1969) found that the prevalence and severity of emphysema was greater in smokers, but, for all degrees of smoking, emphysema was worse in residents of St. Louis than those in less polluted Winnipeg, Canada.

Because the accumulation of particulate matter typically results in pigmentation in lungs, several studies have evaluated the pigmentation and particle deposition of lungs in urban air. Pratt and Kilburn (1971) examined the degree of lung pigmentation and found it was affected by both smoking and urban pollution. Another study found dust (Sweet et al., 1978) and accumulation in urban dwellers. These studies provide evidence for the longer lifetime of soluble particles in the alveolar region.

These autopsy studies, particularly the more recent work in Table B-9, suggest that deposition of ambient coarse particles in the alveolar region is a matter of some concern. Unfortunately, they provide no basis for reliable quantitative assessment of potential health effects or risk factors.

E. Cancer

The relationship between particulate air pollution and cancer has been approached in essentially three ways:

1) Whole animal (*in vivo*) and or *in vitro* tests of collected ambient particulate and extracts;
2) Animal and occupational epidemiologic studies of specific substances found in ambient particles; and
3) Retrospective community epidemiology.

1. Toxicological Studies of Ambient Particles

A number of early studies examined the carcinogenicity of combustion emissions, ambient air and extracts (e.g. Campbell, 1939; Leiter et al., 1942; Kotin et al., 1954). These studies have generally found that organic extracts were more potent tumor producers or promoters than whole samples and that, based on the size distribution of such organics, much of the tumor producing activity should be found in the fine fraction. A notable exception is the observation of McDonald and Woodhouse (1942), who found dust from a freshly tarred road was carcinogenic to animals. Early studies of extracts of particles from atmospheres dominated by coal combustion and industrial emissions also found the aromatic (neutral) fraction of organics dominate tumorogenicity (Stern, 1968; Wynder and Hoffman; 1965). This fraction contains polycyclic aromatics such as BaP.

More recent studies on current U.S. aerosols have focused on the mutagenic potential of airborne particles as indicated by Ames and other *in vitro* tests (Pitts *et al*., 1977; Huisingh, 1981; Daisey, 1980). Mutagenic extracts have been found by preliminary studies in a number of urban areas and, to a lesser extent, non-urban particles (Kolber *et al*., 1981; Pitts *et al*., 1978). As in earlier tumorogenesis studies, organic extracts and smaller particles tend to dominate activity. The neutral fraction, however, does not appear to be most potent on a unit-mass basis (e.g. Figure B-2). Moreover, the coarse fraction contains some mutagenic activity (Kolber *et al*., 1981). These animal and *in vitro* studies of particles are useful in identifying potential problems but are limited by sampling artifacts, questions on the bioavailability of the extracts, and their relationship to human exposures.

2. Studies of Specific Particulate Carcinogens

A number of particulate substances, normally present in ambient air in trace amounts have been found to be carcinogenic in animal and occupational studies. Examples include several polycyclic organics, compounds of trace elements such as chromium, arsenic, nickel, and beryllium and minerals (e.g. asbestos) (See NIOSH, NAS documents on these substances). Inorganic

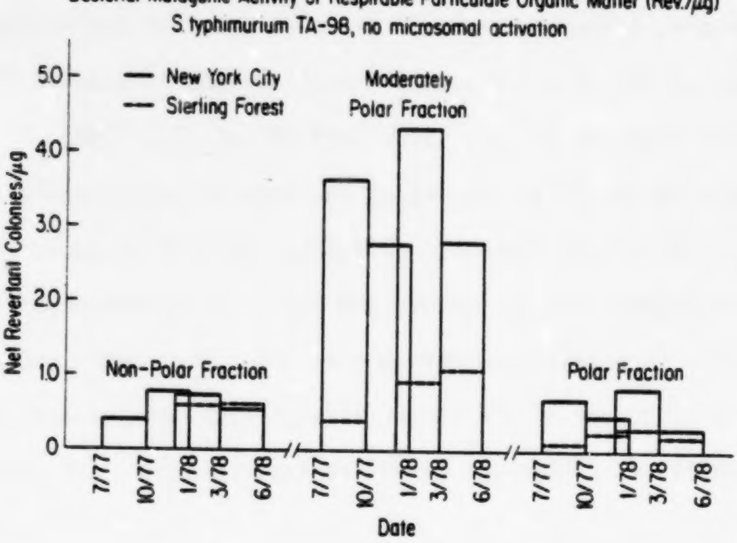

Figure B-2. Bacterial mutagenicity of three organic fractions of size specific particulate matter (<3.5 μm) collected in New York City and Sterling Forest (The latter site is in Tuxedo, N.Y., about 70 km northwest of Manhattan, Daisey, 1980). The moderately polar fraction is a more potent direct acting mutagen than the PAH containing neutral fraction. All fractions show somewhat more activity in the winter. The polar fraction accounts for over half of the organic mass followed by the non-polar and polar fraction respectively (Daisey et al., 1980).

materials (iron ore, carbon, asbestos) have been found to potentiate the oncogenic effect of BaP in intratracheal instillations in rodents (Pylev and Shabad, 1973; Stenback et al., 1976; Saffioti et al., 1968).

These insoluble particles may act as carriers for carcinogens (Stenback et al., 1973) or delay lung clearance (Hilding, 1957) which would increase the contact time between the carcinogen and bronchial mucous membranes. This is especially important considering that most lung tumors occur at the carina or bifurcations of the larger airways (Schlesinger and Lippmann, 1978) where deposition is high and particles tend to accumulate (see Section V-A).

3. Community Epidemiology

Due to the apparent lag between exposure and onset of cancer, available community epidemiological evidence reflects more the polluted conditions in U.S. and British cities in the 1950's and 1960's. The data indicate that cigarette smoking is the dominant cause of respiratory cancer, but that some carcinogenic risk related to air pollution may be present. A higher incidence of lung cancer morbidity and mortality among smokers and non-smokers in urban compared with rural areas, suggests that differences in smoking habits and specific occupational exposures cannot account for all of the urban excess in cancer rates. Several observations implicate particulate matter as contributing to this urban/rural gradient.

As evaluated in several review articles (Higgins, 1976; Holland et al., 1979; Doll, 1978), reductions in particulate matter in London (1954-70) were followed by a reduction in male lung cancer mortality in that city for 1960-1970 (Higgins, 1976). Lung cancer mortality in other urban areas remained constant and increased in rural areas. Although changes in cigarette tar content or consumption may have been responsible, Higgins (1976) concludes that reductions in ambient smoke levels may have been partly responsible. Support is provided by cross-sectional studies that have found weak associations between BS levels and lung cancer in England, after adjusting for cigarette smoking (Higgins, 1976).

Analyses of differential cancer rates between areas are usually limited by a lack of social class, occupational, smoking and mobility information, indirect estimates of exposure to air pollutants, and the long induction time of cancer. Associations between lung cancer and specific indicators (BaP) (NAS, 1972) are not reliable in the context of current exposures. These difficulties preclude any quantitative relationships to be drawn from community studies of particulate matter and cancer.

d) Summary of Evidence on Carcinogeneses

Cigarette smoking generally is considered to be the major determinant of lung cancer (Doll, 1978). The high particulate air pollution of the 1940-60's contained carcinogens and may have potentiated or otherwise contributed to elevated cancer rates in urban areas. Based on filter chemical analyses, levels of polycyclic organics and some inorganic carcinogens have declined since that time and hence carcinogenic risk from these substances has been reduced (Section IV). Studies of current U.S. air suggest mutagenicity of particulate extracts is dominated by other organic particle fractions that may not have been of significance in historical settings. Epidemiological evidence relating to current exposures will not be available for years.

In essence, available evidence does not unequivocally show that current particle exposures contribute to cancer. Neither do they disprove any effect. Because lung cancer is the leading cause of cancer-related deaths, the risk of a small effect could assume some importance. The presence of mutagens in organic particulate fractions from unidentified sources and the potential interaction between these or other particles and carcinogens from cigarettes or occupational exposures suggest some need for caution and further study.

F. Mortality

1. Acute Studies

Community epidemiological studies linking acute particle exposure to mortality are listed in Table B-10. During the "killer fog" episodes in Europe and the U.S., mortality rates rose far above their expected levels, coincident with several days of extremely high levels of particulate and sulfur oxide pollution presumably (Firket, 1936; Schrenk, 1949; Ministry of Health, 1954). Subsequent deaths and chronic disease attributed to severe illnesses during one such episode has also been reported (Ciocco and Thompson, 1961).

Other episodes, though less dramatic, have been reported in London (see CD, Table 14-1) and New York City (Greenburg et al., 1962, 1967). As noted in Section 14.3 of the criteria document much smaller estimates of excess mortality were reported for the New York episodes (at most 4 to 20%) compared to the 15 to 350% death rate increases during the London episodes. Several factors could account for these differences, for example: 1) extremely dense fog associated with the higher levels of sulfuric acid in London (see Section IV); 2) differences in diurnal patterns of pollution levels and in

TABLE B-10. EVIDENCE FROM EPIDEMIOLOGICAL STUDIES SHOWING QUALITATIVE ASSOCIATION BETWEEN SHORT-TERM CHANGES IN MORTALITY AND EXPOSURE TO PARTICLES

General Pollution Characteristics	Observed Effects/Comments	Reference
Severe air pollution episodes lasting several days. Meuse Valley, Donora, London episodes characterized by dense fog, prolonged continuous exposures. NYC episodes had less fog, more diurnal variation in pollution. Complex pollution mix dominated by emissions from fossil fuel combustion, industrial processes, other urban sources and characterized by sudden increases in PM and SO_2 to extremely high estimated levels.	Increased mortality and/or illness, especially among individuals with preexisting cardio-respiratory disease (e.g., bronchitis, emphysema, cardio-pulmonary disorders). Mortality rates appear lower in NYC than in other area episodes. Available autopsies (Schrenk, Min. of Health) generally revealed non-specific irritations of upper respiratory tract. Imprecise aerometry but clear qualitative associations.	Firket, 1931 Schrenk _et al._, 1949 Ministry of Health, 1954 Scott, 1963 Glasser _et al._, 1967
10-year follow-up of Donora episode.	Survivors of Donora episode with prior chronic disease, and those who became acutely ill during episode had higher subsequent mortality and illness.	Ciocco and Thompson, 1961
Minor pollution episodes during London winters when emissions from domestic coal combustion were high, characterized by periodic increases in PM (BS) and SO_2 to high levels.	Increase in daily mortality above 15-day moving average, especially among individuals with preexisting cardio-respiratory disease. Bronchitis, but not pneumonia, mortality significantly correlated with pollution. Peak mortality occurred during an influenza epidemic on a day with peak pollution levels. Meteorological variables do not confound associations.	Martin and Bradley, 1960 Martin, 1964
Daily fluctuation during 14 winters (1958-1972) in London pollution mix over a period when PM (BS) and SO_2 levels declined by 80% and 50%, respectively.	Clear association between daily mortality and daily pollution across the 14 winters, primarily due to smoke and not SO_2. Advanced analysis with superior data base. Standard errors of regression coefficients liable to be large. Possible over-adjustment for weather. Demographic changes not considered. Quartile analysis separating SO_2 and smoke is preliminary and questionable.	Mazumdar _et al._, 1981
Daily fluctuations during 5 winters (1960-1964) in NYC pollution mix containing emissions from fossil fuel combustion, industrial processes, and vehicular traffic and characterized by variations in PM (measured by CoH or smokeshade) and SO_2.	Apparent increase in daily mortality above 15-day moving average and 5-year daily average on days with high CoH and SO_2 levels. SO_2 most strongly correlated with mortality. Data from one central monitoring station used to estimate city-wide exposure levels. Mortality and temperature, but not other environmental variables, adjusted. Some internal inconsistencies, relationship clear only for 1963. See follow-up NYC analyses below.	Glasser and Greenburg, 1971
Daily fluctuations during 14 winters (1963-1976) in NYC pollution mix (see above) over a period when PM and SO_2 levels declined.	Consistent association between smokeshade and daily mortality. Reduction in SO_2 levels were not accompanied by decreased mortality rates. SO_2 not significant in multiple regression. Temperature strongly correlated with mortality. Data from one central monitoring station used to estimate city-wide exposure levels. Demographic changes not considered.	Schimmel and Murawski, (1976) Schimmel, (1978)
Daily fluctuations during 10 years (1962-1972) in NYC pollution mix (see above).		Buechley, 1975
Daily fluctuations during 5 winters (1972-1977) in Pittsburgh, Pa., characterized by variations in PM (CoH) and SO_2, three monitoring sites.	Clear association between daily mortality and PM but not SO_2. Seasonal and temperature effects controlled. Preliminary results from ongoing study. Improved exposure estimates but complex terrain limits for quantitative purposes.	Mazumdar and Sussman, 1981
Daily fluctuations in complex urban air pollution mix in NYC (1962-1965). Philadelphia (1963-1964), L.A. (1962-1965) and Tokyo (1966-1969).	Consistent relationships between 5 periods of high pollution and increases in deaths. Temperature adjusted. Inadequate aerometry.	Lebowitz, 1973

the duration of inversions between the two cities, and; 3) differences in exposure patterns, e.g., London residents typically were exposed to higher levels of indoor pollution.

Studies of two London winters, one being particularly foggy, have shown consistent relationships between particles in the presence of SO_2, and daily variations in mortality (Martin and Bradley, 1960; Martin, 1964). These studies, and reanalyses by Ware _et al_. (1981) are discussed more fully in Sections V-D and VI-C. An exploration of the relationship between daily levels of pollution to daily mortality in London, extended to 14 winters, 1958-59 to 1971-72, (Mazumdar _et al_., 1981) is also discussed in these sections.

The attempt of Mazumdar _et al_. (1981) to separate SO_2 and particles is of particular interest. Based on quartile and multiple regression analyses, a strong association between daily mortality and daily pollution, primarily smoke and not SO_2, was found for 1) all days in the data set, 2) episodic days (days when smoke values exceeded 500 $\mu g/m^3$ plus the seven days adjacent on either side), and 3) non-episodic days (the remaining days). No synergistic effects between the pollutants could be shown.

Advanced analytical techniques were applied in this study in an attempt at separating the effects of SO_2 from smoke and to eliminate the confounding influences of meteorological variables, seasonal and yearly trends, and the strong correlation between the two pollutants. The complexity of the mathematical approach, however, requires cautious interpretation of the results. For instance, there may have been over-adjustment for weather influences, mortality being regressed on seven weather variables. Therefore, it is not clear whether the effects of SO_2 and BS were truly separated. Despite the limitations, the study's superior data base, the attempt at specifying the most appropriate model, and the internal consistency of results add validity to the analyses. The ongoing year by year analyses of this data base should provide additional insights into the potential relationships.

The series of studies that analyzed daily mortality variations in New York City from 1960-1976 also attempted to separate the effects of SO_2 from particulate pollution by calculating the partial correlation coefficients between individual pollutants and health measures. Glasser and Greenburg (1971) reported that SO_2, (based on hourly readings from a single monitoring station) was associated most strongly with daily mortality during the 5 year period, 1960 to 1964. Although the analysis was adjusted for temperature and seasonal variability in mortality rates, no adjustment was made for other environmental variables.

Schimmel and Murawski (1976) and Schimmel (1978) analyzed daily New York City mortality data using time series techniques (e.g., "pre-filtering, nested quartile analysis), refined subsequently by Mazumdar _et al_. (1981). They concluded that particles, as measured by coefficient of haze (CoHs), were associated with mortality and that SO_2 is merely an index variable for other unmeasured extraneous variables and is not a cause of mortality. Buechley (1975) employed similar techniques for approximately the same time period and reached similar conclusions after allowing for many factors (e.g., heat waves, annual mortality cycles, influenza epidemics). This approach, however, unlike Schimmel's, depends upon the specification of the adjustment variables and cannot avoid confounding between the air pollution variables and other components of mortality.

These analyses, like Mazumdar et al. (1981), represent methodological advances over similar epidemiological studies, particularly in their explicit recognition of the time series structure of the data and the requirements this imposes on analysis and conclusions. A number of limitations should be recognized, however, for example: 1) the effect of temperature which was great in NYC, in contrast to London, was treated by a linear analysis, although a nonlinear treatment might have been more appropriate, 2) as in the London study (Mazumdar et al., 1981), changes in the demographic profile in the New York City population during the study period were not considered, 3) as in the London study, the many coefficients studied and the intercorrelations among variables make the standard errors of the coefficients liable to be large, and 4) it is doubtful that measurements from a single monitoring station in central New York City could generate representative exposure estimates for the entire metropolitan area (Goldstein and Landovitz, 1975; 1977a; 1977b). In contrast, Mazumdar et al. (1981), like Martin and Bradley (1960), used pollution data measured at seven stations, which represented the London metropolitan area relatively well (CD, p. 14-19).

Based on a preliminary time-series analyses similar to that of Mazumdar et al. (1981), Mazumdar and Sussman (1981) found that daily levels of particulates (as measured by CoH at three monitoring stations), but not SO_2, were associated with daily mortality in Pittsburgh, Pennsylvania. Another approach was developed by Lebowitz (1973) who, using a stimulus-response model, showed that daily mortality in New York (1962-1965), Philadelphia (1963-1964), Los Angeles (1962-1965) and Tokyo (1966-1969) was associated with high concentrations of SO_2 and particles as measured by TSP (Los Angeles, Philadelphia) and by CoH (New York, Tokyo).

In summary, these acute studies, both episodic and time-series, indicate that high levels of particulate matter, in combination with SO_2 increase mortality rates among individuals with cardiorespiratory disease and influenza. The analyses of daily mortality in London show that the associations may be stronger for particles in some cases, however, SO_2 cannot be clearly excluded. The evidence from New York City analyses is less clear, mainly because of inadequate aerometric data and the confounding influences of temperature.

2. Long-Term Regression Analyses

A number of retrospective regressions analyses have examined the relationship between mortality and sulfur-containing particulate matter as indicated by sulfates. These studies have assumed some prominence in policy analyses and risk assessment of coal combustion (e.g., Wilson et al., 1980). A listing of cross-sectional geographic comparisons is given in Table B-11.

The most widely quoted of these studies are the analyses of Lave and Seskin (1977), which have consistently indicated an association between both sulfates and particulates and mortality. They failed to find a consistent association between non-pollution related effects, such as suicides, venereal diseases, and crime, and sulfates or TSP and hence concluded that the pollution variables were not acting as surrogates. The study's validity in demonstrating a causal association between mortality and TSP and sulfate is limited because of several factors, for example: 1) the omission of important variables (e.g., smoking, diet), 2) aggregate analysis of inadequate data (e.g., minimum sulfate levels), 3) estimation methods which possibly make inefficient use of the information

TABLE B-11. EVIDENCE FROM RETROSPECTIVE REGRESSION ANALYSES RELATING MORTALITY RATES TO CHRONIC SULFUR-CONTAINING PARTICLE EXPOSURE

Source of Data	Elasticity*	Findings/Comments	Reference
Regression using mortality data from 112-117 U.S. SMSAs (1960-64, 1969, 1971) and pollution data (TSP, SO_4 from 1960 and 1970)	≈1.0 for SO_4 & TSP .50-.59 for SO_4 .44-.56 for TSP	Pollution described 8% of total mortality after accounting for urban factor (pop. density, income nonwhite %, >65 yr. old pop.) After limited control for occupational exposures, climate and home heating characteristics, residual pollution effects remained. Did not distinguish TSP and SO_4 effects. Inadequate, complex pollution data (e.g., minimum sulfate levels may have overestimated effects by factor of 2). No control for smoking, diet, other env. variables. Crude adjustments for age and socio-economic status. Internal inconsistencies.	Lave and Seskin, 1977
Regression using mortality and daily pollution (TSP, SO_4) data from 96 U.S. SMSAs, counties, and cities (1974).	1.32 for SO_4 0.06 for TSP (not significant)	Strong, consistent association reported between sulfates and mortality; particulate variables not significant in contrast with Lave and Seskin. Alternate specifications of model and geographic units tested. Simultaneous equation model had little impact on coefficients in contrast to Gerking and Schulze (see below). Smoking, alcohol consumption, industry and occupational mix, medical care, and nutrition variables included. Use of crude surrogates and inadequate aerometric data precludes consistent conclusions.	Chappie and Lave, 1981
Replications using parts of same data base Lave and Seskin used	————	Similar findings, although magnitude of elasticity decreased. Regression coefficients found to be unstable (Thibodeau).	Thibodeau _et al_., 1980 Christainsen and Degen, 1980
Regression using 46 U.S. SMSAs, (1959-61) and TSP, SO_4, SO_2 emissions	.22-.45 for SO_4 plus SO_2	Significant association reported between SO_4 levels and SO_2 emissions and mortality. Negative association between mortality and SO_4. Ridge regression analysis may be biased - cannot test for statistical significance. Pollution coefficients constrained to positive values in least squares analysis - may be unreasonable. Smoking, climate, socio-economic status and occupational parameters included in data. Crude surrogates, inadequate aerometric data used.	Schwing and MacDonald, 1976

TABLE B-11. EVIDENCE FROM RETROSPECTIVE REGRESSION ANALYSES RELATING MORTALITY RATES TO CHRONIC SULFUR-CONTAINING PARTICLE EXPOSURE
(CONTINUED)

Source of Data	Elasticity*	Findings/Comments	Reference
Regression using mortality data from 60 U.S. cities (1970-1976) and SO_2, TSP and NO_2 annual averages	.01 for combined pollutants/total mortality (not significant)	TSP correlated with mortality pneumonia and influenza; SO_2 correlated with infant diseases. Model can't separate influences of medical care availability and pollution exposures. Use of median age poor indicator. SO_2 not as good an indicator as SO_4 - local distributions. Crude surrogates used, inadequate aerometric data used.	Crocker *et al.*, 1979
	—	Reproduced Lave and Seskin model. Nonsignificant pollution coefficients after adding smoking, nutrition, exposure to cold, and medical care variables and using simultaneous equation methods which attempts to control for intercorrelations of variables. Crude surrogates, inadequate aerometric data used.	Gerking and Schulze, 1981
Regression using mortality data from 60-181 U.S. cities and 60 SMSAs(1969) and TSP, SO_4 manganese, iron, benzopyrene annual averages.	≈.4-1.0 for combined pollutants. Significance varied with different model specifications.	TSP and manganese correlated with mortality. Inconsistent correlations with SO_4. When smoking variable added, elasticity reduced by one-half. When only cities analyzed, effects of sulfates possibly ignored. Crude surrogates, inadequate aerometric data used.	Lipfert, 1980
Regression using 404 U.S. counties (1970) and TSP, SO_4, SO_2, CO annual averages	0.3 for SO_4	SO_4 correlated with mortality. TSP correlated with heart failure; SO_2 with lung failure. Linear dose-response relationship; no threshold. Cigarette smoking, migration indirectly included in analysis. Daily extremes in pollution had no effect on annual mortality rate. Crude surrogates, inadequate aerometric data used.	Mendelsohn and Orcutt, 1979

*Elasticity-Expected % change in mortality associated with 10% increase in pollutant from mean value.

contained in the data, 4) the failure to consider inherent simultaneous relationships, making interpretation of the estimated coefficients difficult, or even meaningless, and 5) internal inconsistencies that were not adequately addressed. (See CD, p. 14-36 to 14-43).

The original Lave and Seskin model was refined and re-estimated using 1974 data (Chappie and Lave, 1981) and although the results were qualitatively similar to those for 1960 and 1964, the particulate variables were not significant. A consistent and statistically significant association between sulfates and mortality persisted. The authors attempted to respond to past criticisms by adding variables for smoking, alcohol consumption, medical care and nutrition, by using city or county instead of SMSA data, by using a generalized least squares estimator instead of ordinary least squares, and by testing a simultaneous equation framework. However, the major criticism of their results owing to the inherent limitations in the mortality and exposure data bases, cannot be removed.

Some cross-sectional regression analyses and critiques of Lave and Seskin's work, have found similar relationships (Thibodeau et al., 1980; Christainsen and Degen, 1980; Schwing and MacDonald, 1976; Mendelson and Orcutt, 1979). Others have demonstrated a much smaller influence of sulfur-containing particles and other particle pollutant indicators (Crocker et al., 1979; Lipfert, 1980), while Gerking and Schulze (1981) found a negative, though non-significant, association.

In general, like any epidemiological studies, these models can only be approximately correct; the surrogate explanatory variables can never lead to an adequately adjusted analysis, and separating associations between mortality rates and pollutant and confounding variables is impossible (Ware et al., 1981). Thus, these studies deal with unknown levels of exposure of ill-defined groups of individuals to unspecified pollutants for unstated periods of time, and fail to control for many variables known to affect health status. As such, this group of cross-sectional mortality studies, although suggestive, do not provide reliable quantitative evidence on effects of particulate matter and sulfur oxides. They are, however, not inconsistent with the evidence from acute studies which suggests that a linkage between mortality and pollution may occur at levels below these observed in the more extreme episodes.

APPENDIX C. VISIBILITY--EFFECTS, MECHANISMS AND QUANTITATIVE RELATIONSHIPS

This appendix discusses and evaluates the key studies in the criteria document that provide evidence on the effects associated with reduced visibility, and on both mechanisms and quantitative relationships linking reduced visibility to atmospheric particles.

A. Evaluation of Visibility

Visibility impairment may adversely affect public welfare in essentially two areas: 1) the subjective enjoyment of the environment (aesthetics, personal comfort and well-being); and 2) transportation operations. As discussed in the criteria document, the aesthetic aspects of visibility values can be categorized according to: 1) social/political criteria, community opinions and attitudes held in common about visibility; 2) economic criteria, the dollar cost/benefit associated with visibility; and 3) psychological criteria, the individual needs and benefits resulting from visibility. These categories are not exclusive, but relate to different approaches for measuring somewhat intangible values.

1. Aesthetic Effects

Assessment of the social, economic and psychological value of various levels of visibility is difficult. The criteria document, an EPA report to Congress (EPA, 1979), Rowe and Chestnut (1981), and Fox, Loomis, and Greene (1979) discuss and evaluate several approaches that have been used or proposed towards this end. In particular, the criteria document indicates that preliminary studies of social awareness/perception and the economic value of visibility in urban and non-urban areas support the notion that visibility is an important value in both settings.

Early social awareness studies (Schusky, 1966; DHEW, 1969, p. 100-102; Wall, 1973) conducted in polluted urban areas have generally shown that at higher pollution levels an increasing portion of the population is aware of air pollution and considers it a nuisance. In St. Louis, a linear relationship was observed between annual particle levels (50 - 200 $\mu g/m^3$ TSP) and public awareness and concern. At 80 $\mu g/m^3$ annual geometric mean TSP, about 10% of those surveyed reported air pollution as a nuisance (Schusky, 1966). Although it is reasonable to attribute more of these and other perception results to particulate matter than to gaseous pollutants (Barker, 1976; Wall, 1973), the relative importance of visibility degradation by plumes and haze as compared to dustfall was not clearly addressed in these studies. A more recent study of perception of air pollution in Los Angeles (Flachsbart and Phillips, 1980) represents the most comprehensive evaluation of major pollutant indicators and perception to date. Five gaseous pollutants (O_3, CO, NO_2, NO, SO_2), 4 particle related indices (TSP, dustfall, CoH, visibility), and 6 averaging times were compared with perceived air quality as reported by 475 respondents living in 22 residential areas in Los Angeles County. Only two indices, visibility and ozone, were consistently significantly related ($\alpha = 0.001$) to perceived air quality for all averaging times. The highest correlation coefficient (Kendall's τ) occurred for yearly visibility ($\tau = -.29$) and for number of days visibility was less than 3 miles ($\tau = 0.32$).* Of the other particle indices, only monthly average dustfall was significantly correlated ($\tau = .12$) with perception. Consistent with other studies, the survey also found that air quality is valued less by minority groups and long-time urban residents than by whites (all income classes) and those with some history of rural residence.

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*Kendall's τ is a non-parametric statistic. The negative correlation between the number of people perceiving poorer air quality and visibility and the positive correlation with the number of days visibility is less than three miles are consistent with expectations.

The two major approaches to economic valuation of visibility include 1) survey (e.g., iterative bidding) and 2) property value studies. The major published iterative bidding studies of visibility, conducted in the rural southwest and in the Los Angeles area (South Coast Air Basin), are summarized and evaluated in Table C-1. The Four Corners and Lake Powell studies deal only with single sources and visible plumes, while the Farmington and Los Angeles studies address haze. The preliminary nature of these studies makes them useful primarily as qualitative indicators of the economic value of visibility. Among the more important limitations of the published results are the following:

1) None of these studies has measured existence values (benefit of just knowing pristine areas exist, regardless of intent to use them) or options values (wish to preserve the opportunity to view an unimpaired vista). Rowe and Chestnut (1981) suggest that existence values of good visibility in natural settings may be significantly greater than measured activity values.

2) The studies may be subject in varying degrees to methodological problems. The Farmington study, in particular, discovered a number of biases probably related to the credibility of the contingent market. These biases were not always large but show the difficulty of valuing visibility through iterative bidding.

3) Even if the available results could be taken at face value, so few studies have been conducted that results cannot be directly transferred to other areas of the country. For example, it might be expected that

TABLE C-1. SUMMARY RESULTS OF ITERATIVE BIDDING VISIBILITY STUDIES (after Rowe and Chestnut, 1981)

Design Elements/Study (Year)	Four Corners, NM (1972)[1]	Lake Powell, UT/AZ (1975)[2]	Farmington, NM (1977)[3]	South Coast Air Basin CA (1978)[4]
Total Interviews Conducted (Percent Usable)	1,099 (69%)	104 (79%)	130 (92%)	345 (NR)
Scenarios	Emissions, strip mining, transmission lines, single power plant A - worst B - middle C - best	Air Quality and Power Plant A - No plant B - Plant, no plume C - Plant with Plume	Air Quality and Power Plant A - Visibility 120 km B - Visibility 80 km C - Visibility 44 km	Air Quality and Health A - Poor Visibility- 2 miles B - Fair Visibility- 12 miles C - Good Visibility- 28 miles
Payment Vehicle	User fees/Electricity Bill	User Fees	Utility Bill/Payroll Reduction User Fee	Utility Bill/Monthly Payments to Conservation Fund
Willingness to Pay Bid Comparisons a) Yearly bids for individual residents households	A-C* $85 B-C $50 A-B -	--	A-D* $82 A-B $57 B-D $43	A-C* $245 B-C $243 A-B $174
b) Daily bid for individual recreationists min. $1.00	--	A-C $2.95	A-D $2.44	--
c) Aggregate Yearly Value	A-B $15.54 million A-C $24.57 million	A-B $.414 million A-C $.727 million	A-B $1.47 million A-C $1.99 million A-D $2.14 million B-C $1.1 million	30% improvement $.58 - .65 billion
Tested for bias/found biases	No/No	No/No	Yes/Yes	Yes/Yes
Comments on Total Values	Values for total affected four corners region; attributed mostly to particulate air pollution reduction at single source, but difficult to separate from other visible factors due to unstandardized scenarios. Measured activity values only.	Examined one of fifteen potentially affected parks; pictoral representations not consistent across A-C. Measured activity values only.	Value for San Juan County, New Mexico and Navajo recreation area only. Affected area up to 10 times larger strategic bias not found, but other bias problems with contingent market found. Measured activity values only.	About one-third of benefits are related to aesthetics, two-thirds to health. Results reasonably consistent with companion property value study. Measured user values. Some questionnaire design biases related to aesthetics.

*Capital letters refer to scenarios listed above. Thus "A-C" in the Four Corners case refers to the bids for going from scenario A (worst) to scenario C (best).

1) Randall et al., (1974)
2) Brookshire et al., (1976)
3) Rowe et al., (1980)
4) Brookshire et al., (1979)

willingness to pay for improved visibility in Los Angeles might be greater than that for areas in flat terrain without background hills or mountains.

Despite their limitations, the iterative bidding studies suggest that visibility is of substantial economic value in both urban and natural settings. Although the value of visibility in other areas may vary significantly from that suggested by studies in the rural southwest and Los Angeles, no a priori reason exists to suggest that visibility is of little value in heavily populated eastern urban and rural areas. With respect to recreational settings, of the 23 most heavily used national parks and monuments, 11 were in the East (NPS, 1981). In 1979 over 90 million visits were recorded in all eastern National Park Service managed facilities.

A large number of property value studies related to air quality have been conducted. These have been reviewed by Freeman (1979) and Rowe and Chestnut (1981). Although a variety of air quality indicators have been used, the results of awareness/perception studies strongly suggest visibility plays an important role in air quality related impacts on property values (Rowe and Chestnut, 1981). This contention is best documented in the case of the South Coast Air Basin property value survey (Brookshire et al., 1979). There, the estimated annual benefit of a 25% - 30% improvement in air quality based on property values was about $500 per household. These results are qualitatively similar to the companion iterative bidding study (about $300/household). The bidding study suggests that 22-55% of the bids to improve visibility were related to aesthetic effects. Both the bidding results and the perception study (Flachsbart and Phillips, 1980) conducted in the same area support the possibility of substantial impacts of visibility on Los Angeles area property values.

None of the other published property value studies are accompanied by companion studies that suggest what portion willingness to pay for improved air quality may be due to visibility. Moreover, theoretical problems remain in relating willingness to pay functions from property value differentials (Rowe and Chestnut, 1981). No single study has examined all of the variables that might be important in influencing property values. Because air pollution tends to be a small influence compared to other variables, earlier studies that examined a limited number of variables are particularly suspect. In essence, the available literature suggests that perceived air pollution, and hence visibility, may have tangible effects on property values in urban areas such as Washington, Boston, Los Angeles, and Denver (Rowe and Chestnut); nevertheless, additional theoretical and empirical work is needed before reliable and transferrable quantitative relationships for visibility evaluation can be established.

2. Transportation Effects

Although all forms of transport may be affected by poor visibility (e.g., slowing of highway traffic by anthropogenically induced fog), at current ambient levels, aircraft operations appear to be most sensitive. When visibility drops below 3 miles, FAA safety regulations restrict flight in controlled air spaces to those aircraft and pilots that are certified for operation under Instrument Flight Rules (IFR) (FAA, 1980a). The most severe impact in such cases is usually on non-IFR general aviation aircraft which are, in effect, grounded or forced to search for alternate landing sites. In 1979, there were about 210,000 active general aviation aircraft which accounted for about 84% of total airport operations. Over 23% of all general aviation aircraft had no IFR capability (Schwenk, 1981). Estimates of the percentage of pilots certified for IFR are not available. Commercial, military, and other IFR aircraft operation also may be affected by reduced visibility.

Under IFR conditions, the number of arrivals and departures per hour can be significantly decreased as compared to Visual Flight Rules (VFR) conditions. The effect varies with airport, and in some cases, the visual range cutoff for the most efficient visual approaches (VAPs) may be 5 miles (FAA, 1980b). For example, the performance standard for one configuration at Boston Logan International Airport is 109 operations per hour for VAPs (5 miles), 79 operations per hour for "basic" VFR (3-5 miles), 79 operations per hour for "controllers" visual approach IFR (2-3 miles), and 60 operations per hour for "category I" IFR (2 miles). Thus, depending on airport configuration, schedules, and the extent and duration of haze induced visibility reduction, delays in commercial and other aircraft operations can occur. In large segments of the eastern U.S., midday visibilities less than 3 miles with no obvious natural causes occur 2 - 12% of the days in the summer and 1 - 5% of the time during other seasons (CD, Table 9-5, Figure 9-31). Visibilities less than 5 miles would, of course, be more frequent. Based on typical eastern summertime diurnal cycles in humidity and light scattering (e.g., Ferman et al., 1981), the occurrence of morning visibilities (6 - 8 a.m.) less than 3 - 5 miles would be somewhat greater than for midday visibility, even discounting naturally occurring fog.

Compared with other modes of transport, air travel is generally considered to be safe. It is not, however, riskfree; based on reasonable expectations and the available record, air pollution visibility impairment would tend to increase risks of aircraft operation (U.S. Senate, 1963). Failure to see and avoid objects and obstructions during flight is one of the ten most frequent cause factors for general aviation accidents (FAA, 1978). Another important cause factor is continued VFR flying into adverse weather.

Although such action is normally attributed to errors in judgment (FAA, 1978), in some cases, pilots who by choice or necessity fly in the mixing layer, could fail to distinguish storm fronts or thunderclouds from the prevailing haze until they are upon the adverse weather. The available data in the criteria document do not, however, permit any quantitative assessment of the risks to commercial and general aviation aircraft operation associated with reduced visibility.

The available information on the effects of visibility on transportation suggest that episodic eastern regional haze tends to curtail substantial segments of general aviation aircraft and slow commercial, military, and other IFR operations on the order of 2-12% of the time during the summer. The extent of any delays varies with airport. Reduced visibility may also tend to increase risks associated with aircraft operations in the mixing layer, but quantitative assessments are not available.

B. Mechanisms and Quantitative Relationships

The mechanisms by which atmospheric pollutants degrade perceived visibility are reasonably well understood (Middleton, 1952; Friedlander, 1977). Visibility impairment is the result of light scattering and absorption by the atmospheric aerosol (particles and gases). The "extinction" or attenuation coefficient (σ_{ext}) is a measure of aerosol optical properties and is the sum of blue sky or Rayleigh scattering by air molecules (σ_{Rg}), absorption by pollutant gases (σ_{ag}), and particle scattering (σ_{sp}) and absorption (σ_{ap}). Visibility is inversely related to total extinction from these sources. Blue sky scattering is relatively constant and is significant only under relatively pristine conditions. Absorption by pollutant gases, notably NO_2, usually contributes only a small amount to total extinction (CD, p. 9-20). Even brown hazes in Denver and Los Angeles formerly attributed solely to NO_2, are often dominated by particles (Husar and White, 1976; Groblicki et al., 1980). Thus atmospheric extinction and visibility impairment are normally controlled by particulate matter. Important causes include natural sources (fog, dust, forest fires, sea spray and biologic sources) and anthropogenic sources of sulfur oxide, soot and other particles, nitrogen oxides, and volatile organics (EPA, 1979).

Reduction of visual range by particle extinction is normally dominated by fine particles.* The only important exceptions are some naturally occurring phenomena including precipitation, fog, and dust storms, where larger particles control visibility. Theoretical calculations show that extinction/unit mass efficiencies are substantially greater for fine particles in the 0.1 to 2.0 μm size range than for larger particles (Faxvog and Roessler, 1978). For most commonly observed size distributions of particulate matter, the increased extinction efficiency of fine particles results in fine particles accounting for most of total extinction even though they are only a third or so of the total mass of particles (Latimer et al., 1978). This theoretical expectation is borne out by the unique experiment of Patterson and Wagman (1977) where independent measurement of light scattering and particle size distributions verified the importance of fine particles in controlling scattering in New York City. In addition, a number of experiments have found consistently high correlation (.8 to .98) between light scattering and fine mass (CD, Table 9-1).

The relative importance of scattering and absorption as well as the extinction efficiency per unit equibilibrated mass (γ) of fine particles varies with location. On large regional scales, about 80-95% of particle extinction is due to light scattering (Waggoner et al., 1980; Wolff et al., 1980), with the remainder due to absorption. In urban areas absorption may account for up to 50% of particle extinction (Waggoner et al., 1980; Weiss, 1978). The particle scattering efficiency/unit mass varies from about 3 - 5 m^2/g at various sites, with higher values tending to occur in eastern locations (CD, Table 9-1).

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*For purposes of this document, fine particles include those smaller than 2.5 μm AED.

The variations in fine particle extinction noted above are due largely to variations in size, chemical composition and to some extent, relative humidity. Based on theoretical (Faxvog and Roessler, 1978) and empirical (e.g., Trijonis et al., 1978 a,b; Stevens et al, 1980; Groblicki et al., 1980) results, two components, hygroscopic sulfates and elemental carbon, generally tend to be most significant. Sulfate, with associated ammonium, and hydrogen ions and water, often dominates regional fine mass and extinction, particularly in the East, while elemental carbon accounts for most of the particle absorption observed in urban areas. The relative importance of sulfates to extinction depends on relative humidity, both at the site in question and perhaps along the transport path where secondary formation occurs. Project VISTTA found that sulfates formed in dry desert air were of relatively low light scattering efficiency, compared to sulfates apparently formed in more humid conditions in southern California and transported to the desert (Macias et al., 1980). Our understanding of the role of fine organics and nitrates in light scattering is hindered by the lack of reliable data. In the eastern regional haze these components are likely to amount to less than half of the sulfate component, but may dominate scattering in western urban areas such as Denver and Portland (Groblicki et al., 1980; Cooper and Watson, 1979). The remainder of fine mass (soil-related elements, lead and trace species) contributes only a minor amount to extinction in most U.S. atmospheres (Stevens et al., 1978).

Humidity is important to visibility because of the presence of fine hygroscopic aerosols (e.g., sulfates) which tend to absorb atmospheric water and thus increase light scattering. Measurements in several areas suggest that the extinction due to fine particle scattering will increase by a factor of about two as relative humidity is increased from 70 to 90% (Covert et al., 1980). Based on laboratory studies, reduction in humidity from 90% to 70% might not produce corresponding decreases in scattering because of hysteresis (Tang, 1980). In essence, the hysteresis effect means that the aerosol may tend to retain water absorbed at higher humidities even at lower relative humidities. This effect has not yet been demonstrated to occur in the ambient air.

Through the Koschmieder equation, the extinction coefficient, measured or estimated from fine particle levels, may be used to estimate visual range (CD, p. 9-6). The relationship has the general form:

$$V = \frac{K}{\sigma_{ext}} \qquad \text{(C-1)}$$

Where: V = the visual range, the distance at which a large black object is just visible against the sky.

σ_{ext} = total extinction, the sum of light scattering and absorption by air molecules, fine particles, and NO_2 (CD, p. 9-19).

K = a function of the intrinsic target brightness and observer threshold contrast (E). E is a function of the observer and of target size.

Although a number of factors may limit the applicability of this relationship, for homogeneous pollution, reliable extinction measurements, uniform illumination, large dark targets, and moderate visual ranges, agreement between experiment and theory is rather good. The correlation between visual range and the scattering portion of extinction is typically on the order of 0.9 where comparisons have been made (CD, p. 9-9 to 9-10).

This relationship depends, in part, on human perception of contrast as well as target size and brightness. For a typical observer with a reasonable time for observation and large black targets, a "threshold" of 0.02 is commonly assumed and K = 3.9 (CD, p. 9-9). As noted in the criteria document empirical determinations of K have yielded somewhat lower values, ranging from 1.7 to 3.6 for studies discussed in the criteria document (CD, p. 9-9). The most complete analysis (Ferman et al., 1981) reported a value of 3.5 for well mixed periods. The lower values likely arise from higher threshold contrasts, non-ideal targets, (too bright and/or too small), and the exclusion of absorption estimates. The available data also suggest that reported airport visibilities may significantly underestimate standard visual range. Thus, lower values of K may be more appropriate for matching airport data with higher values for observations in natural settings (CD, p. 9-43).

The relationship between extinction due to dry particle scattering and fine mass is sufficiently stable over a wide range of areas that reasonably quantitative estimates of fine/particle visibility relationships can be made where long-term relative humidity is low (<70%) and particle absorption is small or otherwise known. For such purposes, the Koschmieder relationship can be written as:

$$V = \frac{K}{\sigma_{ag} + \sigma_{Rg} + \gamma FMC} \qquad \text{(C-2)}$$

Where FMC = fine mass concentration, and

$\gamma = (\sigma_{ap} + \sigma_{sp})/FMC$

σ_{ap} = particle absorption coefficient (in units of inverse distance, (e.g., km^{-1})

σ_{sp} = particle scattering coefficient

σ_{Rg} = Rayleigh or "blue sky scattering" ($Rg \approx 0.12\ km^{-1}$)

σ_{ag} = absorption by gases (usually small in non-urban areas)

Thus, with appropriate K and γ derived from available studies, visual range can be estimated from fine mass. Although less certain, the measurements of Covert _et al_. (1980) and regression relationships developed by a number of investigators can be used to estimate fine particle/visibility relationships for higher humidities and hygroscopic aerosols. The criteria document indicates that to correct for the humidity effect, γ (as determined by heated nepholometers and equilibrated filters) should be increased by a factor of about 1.5 at 80% RH, and about 2 at 90% RH. The Koschmieder relationship strictly applies for short-term observations. In estimating long-term (e.g., annual) average visibility from long-term fine mass data, the temporal distribution of fine particle concentrations (e.g., lognormal) must be specified, or median values used.

Figure 7-2, adapted from Figure 9-18 of the criteria document, presents fine particle/visual range relationships for three cases selected as representative of the range of normal situations encountered in the eastern U.S. regional haze and in western urban areas:

1) $\gamma = 3\ m^2/g$; representative of a dry aerosol (CD, Table 9-1) at $\leq$ 50% RH. Absorption may be 10% of extinction where σ_{sp}/unit mass = $2.7\ m^2/g$. This is close to typical measurements in western areas but below most eastern data (CD, Table 9-1).

2) $\gamma = 6\ m^2/g$; representative of the same aerosol as in 1) at 90% humidity, σ_{sp} increased by a factor of 2.

3) $\gamma = 10\ m^2/g$, representative of the similar aerosol, but with absorption accounting for 40% of extinction. Such high absorbtion (predominately associated with carbon) is likely only in urban areas.

Each case may actually be representative of a variety of aerosols. For example, case 2 closely approximates the aerosol observed by Ferman et al. (1981) during their month long study in the Blue Ridge Mountains, even though typical daytime humidities were less than 70%. In this study corrected $\gamma = 5.5\ m^2/g$ as measured by heated nephelometer and when the measured effect of condensed water is added, γ increases. Thus, even though 90% RH is comparatively rare during the daylight hours, case 2 is likely to be closer to typical summertime eastern conditions than is case 1.

Figure 7-2 shows the powerful effect of humidity and carbon absorption on visual range for a given particle level. The curves also indicate that visibility becomes more sensitive to changes to fine particle levels below about 100-200 $\mu g/m^3$. Results from this figure should not be compared directly with airport visibility data. Due to non-ideal targets and observation conditions, airport visibilities will tend to be lower than predicted by the Koschmeider relationship with K = 3.9.

APPENDIX D. TERMS USED TO INDICATE PARTICULATE MATTER

A number of terms are used in the staff paper to indicate various fractions of particulate matter. Table D-1 lists the most frequently used terms and briefly summarizes the relevant size range, definition, and measurement principles for each. Terms are grouped according to their discussion in the staff paper. The table is intended as a ready reference for the reader. A more complete listing of terms and discussion of measurement can be found in the criteria document.

Because conversion among various indicators may be site and time specific, conversion factors are not listed in the Table. Approaches for bounding TP levels from BS readings for certain periods of time in London are presented on pages 96-99 (Section VI-C) of the main text. The relationships between TSP and both IP and TP for contemporary US urban areas are summarized on page 19 (Section IV) and pages 108 and 115 (Section VI).

TABLE D-1. TERMS USED IN THE STAFF PAPER TO INDICATE PARTICULATE MATTER
(CD, Chapters 2,3)

Indicator	Size Range (D_{50})*	Definition/Comment	Measurement Principle
Indices used in Aerosol Science (CD, Chapters 2,9)			
Suspended Particle	~ 0.005 - ~ 100 µm	Generic term - solid or liquid subdivision of matter. Size range approximate for particles suspended in the atmosphere.	NA
Aerosol	NA	Generic term - a disperse system with a gas-phase medium and a solid or liquid condensed phase.	NA
Fine Particles	< 2.5 µm	The combination of nuclei mode (< .1 µm) and accumulation mode (0.1-2.5 µm) particles as collected by a sampler with D_{50} of 2.5 µm. This size is in the vicinity of a "natural" minimum in particle mass; the minimum results from different origins of properties of fine and coarse particles (See Section IV). Fine particles are largely derived from condensation of materials produced during combustion or atmospheric gas formations.	Filtration/gravimetric mass determination
Coarse Particles	> 2.5 µm	Those particles greater than 2.5 µm collected by a particle sampler. Largely derived from mechanical processes (e.g., wind erosion, grinding).	Filtration/gravimetric mass determination
Extinction Coefficient	Varies with instrument	A measure of the optical properties of an aerosol as evidenced by light scattering (nephelometer) and absorption. Usually extinction is dominated by fine particles with the absorption component controlled by carbon. Units are inverse distance (e.g., km^{-1})	Total extinction by light transmission. Scattering, absorption components can be measured separately by various techniques
Indices used in Community Epidemiology Studies (CD, Chapter 3)			
Total Suspended Particulate Matter (TSP)	≤ 25-45 µm	The particulate mass collected and retained by the High-Volume Sampler. Upper size varies with windspeed and direction; subject to artifacts (mostly positive). Widely used in US, Canadian studies.	Filtration/gravimetric mass determination
British Smoke (BS)	≤ 4.5 µm	Pseudo-mass indicator related to small particle darkness (i.e. carbon content). Reflectance readings converted to mass (as $\mu g/m^3$) through use of standard curve, but BS/mass relationships are site and time specific. Widely used in British, other European studies.	Filtration/Optical reflectance reading
Coefficient of Haze (CoH)	≤ 5 µm	Similar to smoke measurement, but measures light transmittance and reports readings in CoHs/1000 ft sampled. Used in NYC, a few other US studies.	Filtration/Optical transmittance reading
Dustfall	> 5 µm	Particles that settle into a dustfall container. Collection highly dependent on collector, location, meteorology. Widely used in Japanese studies, frequently reported elsewhere.	Settling in container/ gravimetric mass determination

TABLE D-1. TERMS USED IN THE STAFF PAPER TO INDICATE PARTICULATE MATTER (CONTINUED)

Indicator	Size Range (D_{50})*	Definition/Comment	Measurement Principle
Indices based on respiratory tract penetration (Miller et al., 1979; ISO, 1981)			
Respirable Particles (RSP, MRP)	< 3-5 μm	Those particles capable of penetrating to the non-cileated portions of the lung. Various approaches to measurements, differ somewhat in size range collected; often used in occupational settings.	Inertial separation followed by filtration/gravimetric mass determination. Gravitational setting, optical devices also used.
Inhalable Particles (IP)	< 15 μm	Those particles capable of penetrating the tracheobronchial and alveolar (thoracic) regions of the respiratory tract; upper size (D_{50}) provides an "envelope" for particle penetration data showing 10% of 15 μm particles reach the tracheobronchial region during mouth breathing. Used in EPA IP network.	Inertial separation followed by filtration/gravimetric mass determination.
Thoracic Particles (TP)	< 10 μm	Same as IP, except upper size (D_{50}) approximates penetration efficiency of the respiratory tract (thoracic region) for mouth breathing.	Inertial separation followed filtration/gravimetric mass determination
Particulate Matter, 10 μm (PM_{10})	< 10 μm	More general term for TP. Includes those particles less than a nominal 10 μm with performance criteria related to respiratory tract penetration data.	Same as TP

*"D_{50}" is one measure of the particle acceptance characteristics of a sampler inlet. It represents the particle size (diameter) at which the inlet is 50% efficient. Samplers collect less than 50% of particle mass for sizes larger than the D_{50}, decreasing to zero efficiency as size increases. Samplers collect more than 50% of the mass for smaller sizes.

APPENDIX E. CASAC CLOSURE MEMORANDUM

UNITED STATES ENVIRONMENTAL PROTECTION AGENCY
WASHINGTON, D.C. 20460

January 29, 1982

OFFICE OF
THE ADMINISTRATOR

Subject: CASAC Review and Closure of the OAQPS Staff Paper for Particulate Matter

Sheldon K Friedlander

From : Sheldon K. Friedlander, Chairman
Clean Air Scientific Advisory Committee

To : Anne M. Gorsuch
Administrator

The Clean Air Scientific Advisory Committee (CASAC) recently completed its second and final review of the document entitled Review of the National Ambient Air Quality Standards for Particulate Matter: Assessment of Scientific and Technical Information, OAQPS Staff Paper. The Committee notes with satisfaction the improvements made in the scientific quality and the completeness of the staff paper. It has been modified in accordance with the recommendations made by CASAC in July and November 1981. This document is also consistent in all significant respects with the scientific evidence presented and interpreted in the combined criteria document for sulfur oxides and particulate matter. It has organized the data relevant to the establishment of particulate primary and secondary ambient air quality standards in a logical and compelling way, and the Committee believes that it provides you with the kind and amount of technical guidance that will be needed to make appropriate revisions to the standards.

CASAC has prepared this closure memorandum to inform you more specifically of its major findings and conclusions concerning the various scientific issues and studies discussed in the staff paper. In addition, the Committee's review of the scientific evidence leading to the particulate standard revision leads to a discussion of its own role in the process for setting the standard.

CASAC Conclusions and Recommendations on Major Scientific Issues and Studies Associated With the Development of Revised NAAQS for Particulates

1. Based upon the review of available scientific evidence, a separate general particulate standard remains a reasonable public health policy choice.

2. CASAC reaffirms its initial recommendation of July 1981 to establish a 10 micrometer cut point for a revised primary particulate standard. This recommendation is based upon a recognition of the periodic, and sometimes frequent, tendency of both healthy and sensitive populations to breathe through their mouths and/or oronasally. This practice increases the amount of particulate matter that can penetrate into the thorax because the larger particles are not filtered in the oronasal passages. Deposition of particulates into this region is of special concern to those individuals with pre-existing respiratory problems and children. In addition, the collection of particles of less than 10 micrometer diameter size more closely resembles particles passing into the thoracic region of the human body than the collection of larger sized particles. Furthermore, monitors equipped for a 10 micrometer cut are less wind dependent and can provide a more accurate profile of the contemporary ambient air than samplers which measure total suspended particles.

- CASAC's recommended size cut is also similar to proposals of other scientific associations. For example, 88% of the national members of the Air Quality Committee of the International Standards Organization recently voted for a particulate cut point at 10 micrometers for sampling particles which can deposit in the lungs.

The CASAC recommendation is based upon available scientific data. Other individuals and groups have discussed the possibility of establishing a revised particulate standard at a size cut considerably less than 10 micrometers. However, for the current revision of the standard, the scientific data more readily support a 10 micrometer size cut.

3. CASAC reached several major conclusions concerning the revision of the 24-hour and annual particulate standards. At the upper bound of the proposed ranges of 150-350 $^{u}g/m^3$ for the 24-hour and 55-110 ug/m^3 for the annual averages, detectable health effects occur in the populations evaluated in the epidemiological studies. Since the upper end of these ranges contain little or no margin of safety, it would be appropriate to consider lower values for revising the 24-hour and annual standards. In addition, the stated ranges are based solely on quantitative evidence reported in epidemiological studies. A final decision on a revised standard should also incorporate information generated through controlled human, animal toxicology, and from other less quantitative epidemiological studies discussed in the criteria document.

There is an absence of a clearly definable exposure-response relationship for particles, as amply discussed in the criteria document and the staff paper. In addition, because airborne particles are heterogeneous in composition, the potential toxic effects of individual constituents should be considered in setting the standard. Thus, compared to margins of safety set for pollutants such as ozone and carbon monoxide, where exposure-response relationships are better established and small margins of safety are more justifiable, CASAC believes you should consider a revised standard with a wider margin of safety.

4. The Committee reached general agreement that the annual particulate standard should consist of an arithmetic mean. It is recommended that the 24-hour standard include a statistical form and that the number of exceedences is set in relation to the revised standard level.

5. During the past decade, the link between visibility and fine particle mass concentrations has been convincingly documented. Visibility is a sensitive indicator of accumulated man-made pollutants in the ambient air. The public cares about visibility and is willing to pay something for clean air. However, the quantitative basis for establishing a psychological, economic, transportation or any other welfare cost associated with visibility impairment has not been established. In addition, controls required to achieve a given visibility standard are not known due to the complexities of pollutant transport and transformation.

Defining acceptable levels of visibility is a social/ policy judgment as well as a scientific decision, but science can provide some guidance. The upper end of the 8-25 ug/m^3 range for fine particles (those particles with a diameter size of less than 2.5 micrometers) would tend to maintain the status quo for the eastern United States and some western urban areas, but would permit air quality degradation for large areas in the west including national parks. Also, it is highly uncertain that the recommended thoracic particle ranges for the primary standard will protect visibility. The 8-25 ug/m^3 range for fine particles suggested for visibility protection is a seasonal and spatial average, unlike peak values which will be recommended for the primary standard.

The strongest case for a visibility related standard is one that links emissions of nitrogen oxides and sulfur dioxide with the interrelated aspects of acidic deposition, possible climatological effects, and visibility. Each of these three air quality issues is related to the fine particles which originate both as primary particulate emissions and as secondary aerosols from atmospheric conversions of sulfur dioxide and nitrogen oxides emitted as vapors. In terms of a control strategy to protect public welfare, it may be more efficient to consider a common standard linked to fine particles than to establish a separate set of controls for each of these problems and pollutants.

6. The Committee's evaluation of scientific data and studies in the criteria document and the staff paper lead it to conclude that there is no scientific justification for the establishment of a particulate standard for the specific protection of vegetation.

7. The Committee discussed what effect elimination of a Total Suspended Particulate (TSP) standard would have on the environment. The soiling and nuisance aspects of TSP are essentially local air quality problems because such coarse particles are not transported great distances. This contrasts with visibility or oxidant related problems which are distinctly issues of long range pollution transport. Individuals who serve on the Committee made various recommendations regarding retention or elimination of a secondary standard for TSP, but no clear consensus evolved.

The Process for Setting the Ambient Particulate Standard

In its report of September 21, 1981, CASAC made several major recommendations relating to the process for setting ambient air standards. The Committee is aware that your staff is analyzing its report and is awaiting a response.

A major underlying assumption of the Committee's recommendations was the need to make more explicit the relationship between the scientific evidence in the criteria document and the staff paper and the eventual selection of a numerical level for individual standards. The Committee strongly believes in the need to clarify the standard setting process by identifying the key studies that will shape the determination of a standard. Intensive evaluation of such studies by CASAC and the public will considerably increase your ability to set a scientifically supportable standard.

The Committee is greatly encouraged by your decision to improve the format and content of OAQPS scientific issue staff papers. In the Draft Staff Paper for Particulate Matter key studies are identified and their implications for setting primary and secondary standards are discussed. More importantly, the inclusion of numerical ranges and their supporting rationale enabled the Committee and the public to critically examine the staff's proposed use of the studies. This led to a marked improvement in the quality of the public dialogue concerning the scientific basis for revising the standard. CASAC commends your effort and recommends that all staff papers developed for ambient air standards contain numerical ranges.

CASAC recognizes that your statutory responsibility to set standards requires public health policy judgments in addition to determinations of a strictly scientific nature. While the Committee is willing to further advise you on the particulate standard, we see no need, in view of the already extensive comments provided, to review the proposed particulate standards prior to their publication in the Federal Register. In this instance, the public comment period will provide sufficient opportunity for the Committee to provide any additional comment or review that may be necessary.

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